Principles
of
NUCLEAR
REACTOR
ENGINEERING

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GLASSTONE

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VAN



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June 1956.

Principles of NUCLEAR REACTOR ENGINEERING

by

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Gift of Dr. Donald P. Le

FOREWORD

by

Lewis L. Strauss, Chairman
United States Atomic Energy Commission

The development of nuclear industry in the American industrial pattern requires ever-increasing numbers of engineers with special skills. The demands for such nuclear engineers are a challenge to the educational system of the United States.

To help education meet this challenge, the American Society for Engineering Education's Committee on Atomic Energy Education recommended three years ago that the Atomic Energy Commission sponsor the preparation and publication of an unclassified sourcebook for reactor engineering education, based upon the experience of the Commission's laboratories and the material in their files.

Acting on the recommendation, the Commission engaged Dr. Samuel Glasstone to write this text. The Oak Ridge National Laboratory, which has operated the Oak Ridge School of Reactor Technology since March 1950, has furnished much source material and has carefully reviewed the manuscript. Advice and criticism of the manuscript have been provided by specialists throughout the national atomic energy program.

The AEC Division of Classification has reviewed the manuscript to assure that it complies with present rules for declassification. The data selected, its technical evaluation, and the conclusions in this book are wholly the work of the author and his associates.

The Commission is gratified by the progress of the program launched by the American Society for Engineering Education to work nuclear materials into engineering courses and to develop curricula specifically to train nuclear engineers. It is hoped that this book will contribute to the further advancement of education in nuclear engineering in the United States. In the interest of vigorous world-wide atomic energy development for peace, the Commission hopes that this book will also aid other nations to train engineers in the disciplines of reactor technology.

Lewis L. Strauss

FOREWORD

by

A. M. Weinberg, Research Director
Oak Ridge National Laboratory

Nuclear technology has developed since the early days of 1942, when the nuclear physics of the reactor dominated the technical thinking, to the present time when the central issues revolve around reactor and radiochemical engineering. This development is most gratifying since it is evident that the technology is maturing. Its complete dependence on nuclear physicists is dwindling; its dependence on nuclear engineers — who have a grasp of the entire field — is growing and will continue to grow.

It is on this account that Dr. Glasstone's "Principles of Nuclear Reactor Engineering" is a very welcome addition to the literature of Reactor Science and Technology. Much of the material for the book was supplied by members of the staff at the Oak Ridge National Laboratory. Since most of these contributors lecture on the subject of their contribution at the Oak Ridge School of Reactor Technology, the contents and point of view of the book inevitably reflect the Oak Ridge judgment as to "What is Reactor Engineering." The scope of the discipline is, of course, a real pedagogical question in the development of a brand new technology and Dr. Glasstone's book ought to be very useful in helping delineate the area of Reactor Engineering.

The book was begun as a joint effort of N. F. Lansing and Dr. Glasstone. Subsequently, Dr. W. H Sullivan of Oak Ridge National Laboratory assumed the job of liaison between Dr. Glasstone and the Oak Ridge National Laboratory staff contributors. To the members of the Oak Ridge National Laboratory staff who contributed, and above all to Dr. Glasstone who so deftly drew together, organized, and wrote "Principles of Nuclear Reactor Engineering," the coming generation of nuclear engineers who will use this text as their introduction to the field will always be indebted.

PREFACE

The field of nuclear reactor engineering encompasses the design, construction, and operation of reactors for the release of energy in a continuous and controlled manner by the fission of certain atomic nuclei. The main purpose of this book is to describe the fundamental scientific principles upon which reactor engineering is based. Actually, the principles are not very complicated and are, on the whole, well understood, but in their translation into practice there are encountered various problems of a somewhat novel character due to the peculiar nature of the fission process and its associated radiations. Reactor engineering is largely concerned with the solution of these problems, involving a cooperative effort in which chemical, electrical, electronic, mechanical, and other engineers must work with chemists, mathematicians, metallurgists, physicists, and even biologists.

It is in order to facilitate such cooperation that this book covers a wide range of topics. The objective is to provide an over-all review which will help the individual engineer to understand the relationship of his activities to that of the group as a whole. In this way he will best be able to apply his knowledge and experience in one or more of the familiar fields of engineering to the solution of the special problems arising from the use of atomic energy as a source of power. It should be emphasized that it is the aim of this book to treat the subject of nuclear reactor engineering in breadth rather than in depth. Nearly every chapter is, therefore, to be regarded as an introduction to a particular aspect of reactor design, in which the specialist will require both further knowledge and practical experience.

Although some of the material presented in this book, especially in the early chapters, can be found in other works, its inclusion here is, nevertheless, felt to be justified for two reasons. First, it is desired to make the book as complete as possible, within the limitations of its essential purpose, and, second, many engineers and engineering students would probably be somewhat discouraged if they had to search the standard texts in nuclear physics, reactor theory, etc., in order to obtain the information they need. Those who wish to delve further into the more theoretical aspects of nuclear reactor engineering should find the essential outlines presented here a useful foundation upon which to base their further studies.

Two classes of reader, in particular, have been kept in mind in the prepara-

tion of this book. One is the practicing engineer who wishes to know something of the impact of nuclear energy upon his professional activities. For such a reader the book should serve as a guide; the inclusion of many cross references and a reasonably complete index should help him find his way through the book even if he does not wish to read it systematically. The other class of reader is represented by students in engineering schools at the senior or first-year graduate level. To judge from the experience with graduate classes held at Los Alamos, under the auspices of the College of Engineering, University of New Mexico, the book should provide the basis for a two-semester course in Nuclear Reactor Engineering Principles.

Although the first reactor developments were made by physicists, engineers have played an increasingly important role in the field. One of the consequences of this dual contribution is that a somewhat mixed system of units has come into use in reactor engineering. For example, energies are expressed in both electron volts and in kilowatt-hours; neutron fluxes are stated in terms of the square centimeter and second, but for heat fluxes the corresponding units are the square foot and hour. The weight of natural uranium fuel in a large reactor is invariably quoted in tons, but for a small reactor, using enriched fuel, it is the general practice to express the weight in kilograms. This confusing situation is one which is unlikely to be remedied in the foreseeable future, and the reactor engineer must learn to live with it. One of the aims of the treatment given here is to help him to do so.

In writing this book I have received help from a large number of individuals, so large, in fact, that it is not possible to express my appreciation by mentioning their names here. A considerable volume of material was made available by members of the staff of Oak Ridge National Laboratory and others, particularly for Chapters V through XI, and acknowledgment for this is made in footnotes to these chapters and on the title page. However, much of the information and references supplied were classified, so that it was necessary to substitute material which permitted illustration of the same basic principles without violating security requirements. In this connection, I am especially indebted to Dr. A. F. Owings, Scientific Adviser, A. E. C. Technical Information Service, for calling attention to, and securing for my use, many declassified reports and other documents. Thanks are also due to Dr. S. McLain, Argonne National Laboratory, for generously placing at my disposal the notes of his Reactor Engineering Lectures. The manuscript has been reviewed, with varying degrees of thoroughness, by a number of engineers and scientists, including members of the Committee on Atomic Energy Education of the American Society for Engineering Education. Although it may be somewhat invidious to select the name of one or other of these for special thanks, I wish, nevertheless, to acknowledge the help given me in various matters by J. A. Lane, Director, Reactor Experimental Engineering Division, Oak Ridge National Laboratory.

In addition to the foregoing, I wish to thank N. F. Lansing, Oak Ridge National Laboratory, for preliminary discussions regarding the general organization of the book; Dr. W. H Sullivan, Oak Ridge National Laboratory, who took over the responsibility for liaison between members of the Laboratory and myself; Dr. A. F. Thompson, Chief, A. E. C. Technical Information Service, and his assistant, J. H. Martens, for maintaining pleasant relations by solving administrative problems; C. B. Holmes, Chief, Editorial Branch, Technical Information Service, Oak Ridge, for preparing the illustrations; and Dr. N. E. Bradbury, Director, and Dr. R. C. Smith, Assistant Director, Los Alamos Scientific Laboratory, for their courtesy in making available office, library, and other facilities in the congenial atmosphere of the Laboratory.

SAMUEL GLASSTONE

Los Alamos, N. M.

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Chapter I *

SCOPE OF NUCLEAR REACTOR ENGINEERING

INTRODUCTION

The World's Energy Resources

1.1. The discovery of nuclear fission in 1939 was an event of epochal significance, because it opened up the prospect of an entirely new source of power, utilizing the internal energy of the atomic nucleus. The basic materials that can be used for the release of nuclear energy by fission are the elements uranium and thorium. Minerals containing these elements are widely distributed in the earth's crust, so that, as will be apparent shortly, they represent a very large potential source of power.

1.2. For the past half century fossil fuels, namely, coal, oil, and natural gas, have supplied the major portion of the world's energy requirements. It has long been realized, however, that in the not too distant future these sources of energy will be largely exhausted. At the present time the total power consumption for all countries is about 0.1×10^{18} Btu, i.e., 0.1 Q, per annum.† Taking into account both the steadily growing population and the increasing per capita power demand, it is possible that by the year 2000 the rate of energy utilization for the whole world will be close to 1 Q per annum. The most reliable estimates indicate that the energy content of the coal, oil, gas, and oil shale that can be recovered at no more than twice the present cost amounts to less than 40 Q. This means that within 100 years the economically useful reserves of fossil fuels will be virtually exhausted.

1.3. Even when allowance is made for errors in the foregoing estimates, the conclusion is inevitable that new sources of power must be found during the next 50 years or so if the earth is to support the growing population with some increase in living standards. Two such sources have been considered: solar energy and atomic (or nuclear) energy. The idea of making more direct use of

* Reviewed by R. A. Charpie.

† The symbol Q is used to represent 10^{18} Btu, so that large quantities of energy can be expressed in terms of convenient small numbers; see P. C. Putnam, "Energy in the Future," D. Van Nostrand Co. Inc., 1953. This work is the source of the data presented here.

the sun's energy is very attractive, but so far no satisfactory method has been developed for utilizing it on a large scale except through green plants. On the other hand, with the discovery of fission, nuclear energy appears to be a practical possibility.

1.4. One of the remarkable facts about nuclear energy is the large amount that can be released from a small mass of active (fissionable) material. Thus, the complete fission of one pound of uranium would liberate roughly 3.6×10^{10} Btu of heat; this is equivalent to the energy produced in the combustion of close to 1400 tons of 13,000 Btu/lb coal.*

1.5. The total amount of uranium and thorium in the earth's crust, to a depth of three miles, is very large, possibly something like 10^{12} tons. However, much of this is present in minerals containing such a small proportion of the desired element that extraction would be prohibitively expensive. Assuming, as is probable, that technological advances will reduce the cost of recovery from moderately low-grade ores to not more than \$100 per lb of metal, it has been estimated that the world reserve, based on admittedly incomplete mappings, is 25 million tons of uranium and 1 million tons of thorium.† As will be seen later, it is unlikely that the whole of this material can be utilized economically in fission, but in favorable circumstances perhaps one third might be so used. On the basis of the energy equivalence factor given in the preceding paragraph, the heat that might be obtained by fission is thus

$$\frac{1}{3} \times 26 \times 10^6 \text{ tons} \times 2000 \frac{\text{lb}}{\text{ton}} \times 3.6 \times 10^{10} \frac{\text{Btu}}{\text{lb}} = 624 \times 10^{18} \text{ Btu}$$
$$= 624 \text{ Q}.$$

The possible energy reserve in the form of uranium and thorium is consequently many times greater than that of the fossil fuels and so it would, if properly developed, represent a very significant contribution to the world's power sources.

1.6. It may be noted, in passing, that the production of energy by fission is by no means regarded as the ultimate solution of the problem of making nuclear energy available for practical use. Scientists are intrigued by the possibility of obtaining controllable energy by the fusion of very light nuclei, the process responsible for the energy of the sun and stars. If this could be achieved, water might become the main source of energy, for the fusion, into helium, of the hydrogen nuclei contained in one pound of water should produce (theoretically) close to 3×10^{11} Btu of energy. Even if the fusion process were restricted to deuterium, i.e., heavy hydrogen, since this would probably be easier to achieve, it would still be possible, in principle, to obtain nearly 10^8 Btu/lb of water.

Nuclear Reactors and the Fission Process

1.7. The basically correct statement, in § 1.4, that the heat released by the complete fission of one pound of nuclear fuel is equivalent to that obtainable from 1400 tons of coal (or 300,000 gal of fuel oil), has led to some erroneous conclusions. The fantastic possibility has been envisaged of including in an automobile enough fissionable material, about the size of a pea, to last the life of the vehicle. In order to realize why this is not within the bounds of reality, it is necessary to understand something about the fission process. Although this will be discussed in more detail in later parts of the book, a brief outline will be given here.

* Throughout this book a "ton" refers to a short ton of 2000 lb.
† P. C. Putnam, *op. cit.*, p. 214.

1.8. Fission occurs when a nucleus of the appropriate type captures a subatomic particle, called a neutron. The nucleus then splits into two lighter nuclei (the primary fission products), and at the same time energy is released. Also, in each act of fission neutrons are emitted, two or three on the average, and if one of these at least is captured by another fissionable nucleus, a fission chain reaction with the continuous production of energy becomes possible. The device in which the nuclear fission chain is initiated, maintained, and controlled, so that the accompanying energy is released at a specified rate, is called a *nuclear reactor*.*

1.9. Because of the loss of neutrons, by escape and by capture in various ways which do not lead to fission, a chain reaction can be maintained only if the system exceeds a certain size, referred to as the "critical" size. Although this size has a definite value for a particular system, it can vary over a wide range, depending on the nature and amounts of the materials present and their geometrical configuration. Thus, for a reactor consisting largely of pure fissionable material (uranium-235 or plutonium-239), the critical size has been described, very roughly, as being about "as big as a football." † From the known density of metallic uranium, it can be readily calculated that a reactor of this type contains approximately 100 lb of fissionable material. If the amount is less than this, the fission chain cannot be maintained and there can be no continuous production of energy. However, in other reactors the critical mass may be smaller or larger, depending upon various circumstances. For example, systems using natural uranium metal as the nuclear fuel require several tons of this material before they become critical. At the other extreme, some reactors can operate on a few pounds of fuel.

1.10. Thus, although it is true that one pound of fissionable material can produce energy equivalent to nearly three million times its own weight of coal, the reactor in which the energy release occurs may contain anything from some pounds to several tons of nuclear fuel. The amount of fuel to be consumed for energy production must be present in addition to the critical quantity, since this is the essential minimum. However, too large an excess is not only wasteful, for various reasons, but it makes the reactor more difficult to control. There is, consequently, a practical limit to the energy that can be produced before the

* The term "pile," although less descriptive of modern reactors, is still used to some extent. It had its origin in the fact that the first successful reactor was constructed by piling layers of graphite (some containing uranium) upon one another (§ 13.4).
† W. H. Zinn, *Nucleonics*, **10**, No. 9, 8 (1952).

reactor is "refueled." Nevertheless, an excess of only 10 lb of fissionable material, over and above the critical amount, could produce the equivalent of 100 million kw-hr in the form of heat.

Utilization of Nuclear Fuel

1.11. There is no method known at the present time for utilizing the major portion of the energy of fission except as heat.* Thus, in a sense, a nuclear reactor may be thought of as a kind of furnace. In the production of electricity, for example, the reactor may be regarded as a substitute for the boiler firebox. The boiler is actually a heat exchanger, but the steam (or other vapor) obtained would be used in a conventional turbogenerator system. A general comparison between a coal-burning, steam-electric generating installation and one using a nuclear reactor as a source of heat is shown in Fig. 1.1. The upper portion is a schematic representation of a coal-fired plant, and the lower section indicates

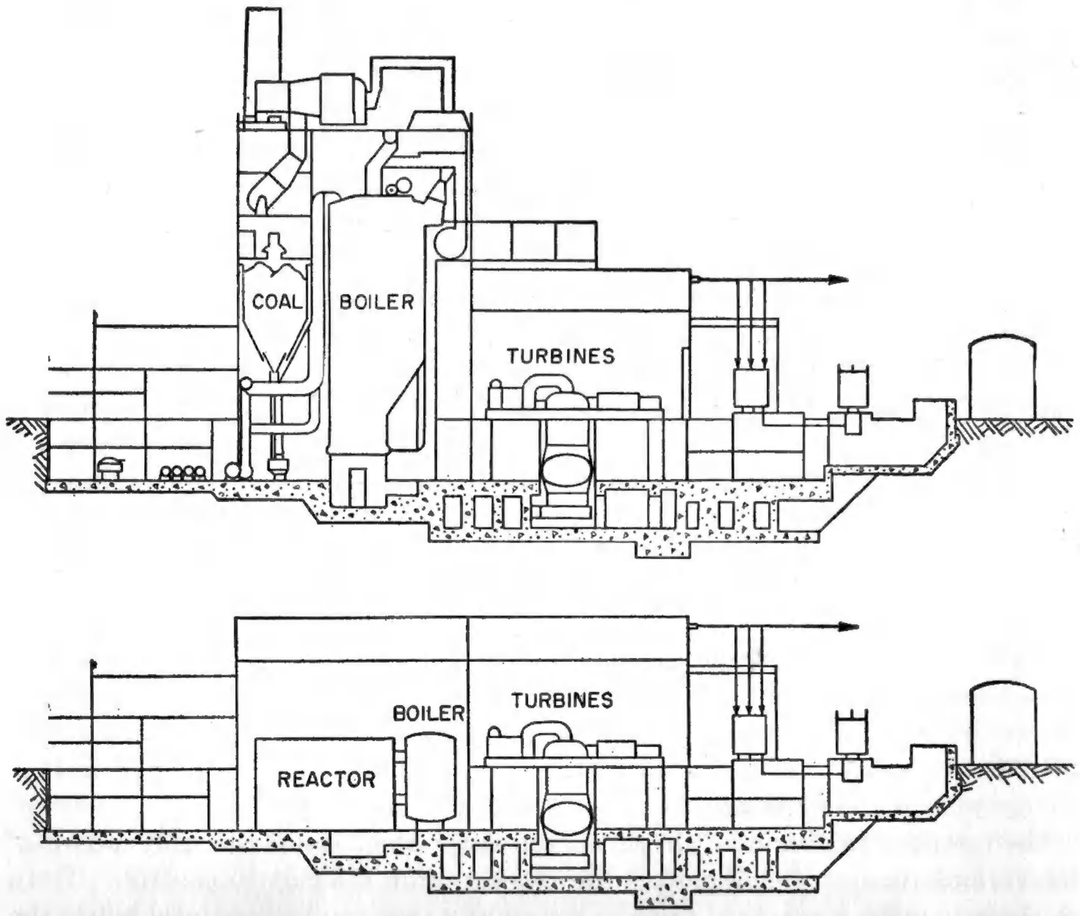


Fig. 1.1. Comparison of coal-fired and nuclear steam-electric power plants (P. Sporn and S. N. Fiola)

*A small fraction of the fission energy, considerably less than one per cent, present as radioactive energy of the fission products, may be converted directly into electrical energy.

the changes that would be necessary if the heat were supplied by a nuclear reactor. The reactor shown is approximately the size of the one at the Brookhaven National Laboratory, including its thick concrete shield (see § 1.31). It is seen that in the nuclear power plant the coal-handling equipment is eliminated and the boiler (or heat exchanger) is smaller than the conventional type. The electrical generating equipment is, however, essentially unchanged.

1.12. There is another aspect of the nuclear reactor problem that may be touched upon briefly here. It was mentioned earlier that uranium and thorium are the basic materials for the production of nuclear energy; this statement requires some amplification. Actually, ordinary thorium cannot be used directly as fuel in a nuclear reactor, and naturally occurring uranium contains only 0.7 per cent of the fissionable species, uranium-235. However, both uranium-238, the main constituent of natural uranium, and thorium can be converted into reactor fuel.

1.13. When uranium-238 and normal thorium (thorium-232) capture neutrons they do not, in general, suffer fission, but yield products that undergo spontaneous changes leading to the formation of fissionable nuclei. In this way, the two inactive species, called *fertile materials*, are converted into different elements, plutonium-239 and uranium-233, respectively, which are fissionable and are capable of acting as nuclear fuels.

1.14. It will be recalled (§ 1.8) that, when a nucleus suffers fission, two or three neutrons are emitted, but one only is required to maintain the chain reaction. Hence, if there were no losses, a neutron (or neutrons) would be available for the conversion of a fertile nucleus into a fissionable one, as described above. As stated earlier, some neutrons are inevitably lost, in one way or another, but it is possible to design a reactor which can produce energy and, at the same time, change fertile material into fuel. It is in this manner that the essentially inactive uranium-238 and thorium-232 occurring in nature can be converted into fissionable material.

Processing of Reactor Fuel

1.15. It would thus appear, at first sight, that since the fissionable nuclei, as they are consumed by fission, can be replaced by a more or less equivalent (or possibly greater) number of other such nuclei, at the expense of inactive fertile material, the reactor would continue to operate without refueling, at least as long as fertile material remained. Unfortunately, this is not the case. In the foregoing argument, the effect of the products of fission — the "ashes" of the nuclear furnace — has been overlooked. As the reactor operates, the fission products, consisting of a considerable number of different nuclei, accumulate. All of these nuclei are able to capture neutrons and so they compete with the fission process. In due course the proportion of neutrons lost in this manner becomes so large that the nuclear fission chain can no longer be sustained.

1.16. Consequently, from time to time, the reactor fuel must be removed and

processed so as to separate the accumulated fission products. In addition, dimensional and structural changes in the fuel units, due to the continued action of various radiations (§ 1.26), may make it necessary to refabricate them from time to time. As a rough indication, it may be assumed that treatment is necessary when 1 per cent of the fertile material has been consumed. This means that the reactor fuel, consisting of both fertile and fissionable nuclei, would have to be processed about a hundred times before all the fertile material could be utilized as a source of energy. Thus, the initial price of less than $100 per lb for natural uranium or thorium, referred to in § 1.5, actually represents a fraction only of the total cost of these elements if they are to be effectively used as nuclear fuels.

Cost of Nuclear Power

1.17. As a result of a failure by certain writers to understand the situation, it has sometimes been implied that extensive utilization of nuclear fission will make available an extremely cheap source of electric power. That this cannot be the case, within the boundaries of present knowledge, follows from the fact that distribution accounts for 65 to 75 per cent of the cost of electricity to the consumer. All that nuclear fission can do, as far as can now be seen, is to replace the fuel, e.g., coal, oil or gas, or water power, in the production of electricity. Consequently, even if there were no charge for the nuclear fuel, the ultimate cost of electric power supplied to the consumer could be reduced by no more than about 15 to 20 per cent.

1.18. It is doubtful whether power from nuclear energy can be generally competitive with electric power from conventional sources during the next 10 or 20 years. However, in due course, as nuclear technology develops, which it undoubtedly will, and fossil fuels become scarcer, it is very probable that nuclear reactors will supply power at a price equal to or less than that from other sources. It should not be forgotten also that, as far as the near future is concerned, there are circumstances when the cost of power is of secondary significance and its availability is of prime importance.

1.19. Such considerations justify the use of nuclear reactors to supply the propulsive power for submarines, naval ships, or military aircraft. Since a nuclear reactor does not require oxygen, a nuclear-powered submarine can remain submerged for long periods. Further, naval vessels and aircraft using nuclear power will be able to travel great distances without refueling, because of the very large energy content of the nuclear fuel. The tremendous saving in the weight of fuel will more than compensate for the mass of the reactor and its accessory equipment.

1.20. The higher cost of nuclear power might be discounted in remote regions where neither coal, oil, nor water is available. In such areas power might be required for the extraction of valuable metals, for example, thus saving the cost of transporting the ore. Remote military bases are now dependent upon tankers

to supply oil fuel; under these conditions nuclear power might be advantageous, especially in wartime when shipping is subject to attack. Finally, nuclear reactors might be justifiable in mobile power stations for use in emergencies or disasters, either in peace or war.

Problems of Reactor Design

1.21. In view of the possible applications of nuclear power, even at the present time, it might be wondered why developments have apparently been so slow. This is not due to any shortage of ideas concerning reactor designs. Because there are a number of variable factors among which a choice may be made, there are many acceptable solutions to the problem of designing a nuclear reactor. However, lack of experience makes it impossible to say which of these will be best for a given set of conditions.

1.22. Consider, for example, the two reactors constructed for use in a submarine, namely, the Submarine Thermal Reactor (STR) and the Submarine Intermediate Reactor (SIR). In the STR the fission chain is maintained by neutrons of relatively low (thermal) energies and the heat is removed by water under pressure. In the SIR, on the other hand, neutrons of intermediate energy cause most of the fissions, and the heat-transfer agent is molten sodium. Each system is known to have both advantages and disadvantages relative to the other, but until both reactors are operated it cannot be said which design will prove to be the better.

1.23. Another illustration of the present uncertainty in choosing an optimum design is provided by the studies made in 1952 by four groups of industrial concerns.* Although all had access to the same information, the designs proposed for a dual-purpose reactor, that is, one which would produce power at a not unreasonable cost and also plutonium for military purposes, differed very markedly.

Problems of Reactor Materials

1.24. The general scientific principles upon which reactor design is based are now well established as will be seen in this book, but there are many problems of a technological nature that have not been completely resolved. It is a well-known principle of thermodynamics that, in order to secure good efficiency, heat should be available at high temperatures; in modern boilers, for example, steam temperatures of 1050°F (565°C) are attained. If such high temperatures

* The four original groups were: Commonwealth Edison Co. and Public Service Co. of Northern Illinois; Dow Chemical Co. and Detroit Edison Co.; Monsanto Chemical Co. and Union Electric Co.; Pacific Gas and Electric Co. and Bechtel Corp. A declassified version of the reports made by these companies has been issued under the title of "Reports to the U. S. Atomic Energy Commission on Nuclear Power Reactor Technology," U. S. Government Printing Office, May, 1953. By the end of 1954, nearly twenty groups, involving a large number of companies, had been approved for participation in nuclear energy studies of interest to industry.

are to be produced in the heat exchanger associated with a nuclear reactor, the latter must be operated above 1100°F. In this vicinity, however, metallic uranium undergoes a phase transition with an accompanying change in density. Reactor fuel elements made of uranium would then change in size and shape, resulting in distortions which cannot be tolerated. Suggestions have been made for the use of molten fuels, either in the form of uranium alloys or of mixed salts, but the handling of such liquids at high temperature is obviously not a simple matter. It is possible that the best over-all economy in the production of nuclear power will be obtained with steam at lower temperatures (and pressures) than in modern, conventional generating plants.

1.25. Materials used in reactor construction must satisfy much more stringent requirements than do those employed in conventional boilers. In addition to great strength, resistance to stress deformation, and ability to withstand high temperatures, reactor materials must not capture neutrons to any appreciable extent. If they do, they will cause serious interference with the propagation of the nuclear-fission chain (§ 1.9). This requirement means not only the use of special materials, but that they may have to be of very high purity. Thus, the relatively uncommon metal zirconium has many qualities which make it valuable in reactor construction; however, in nature, the closely related element hafnium, with a considerable ability to capture neutrons, is invariably associated with zirconium. Unless special steps are taken to obtain the latter free from hafnium, it is, therefore, useless for reactor work.

1.26. Another restriction upon reactor materials, which does not arise in conventional practice, is the ability to withstand the continuous bombardment of various nuclear radiations, including neutrons, present in the reactor. Such radiations can change the internal structure of solids, with the result that their physical and mechanical properties may be seriously affected. Materials must therefore be developed that are not susceptible to what is called "radiation damage."

Heat Removal Problems

1.27. Since the reactor is essentially a device for producing heat, this heat must be continuously removed in some manner. In general, a fluid heat-transfer medium (or coolant) would be circulated through the reactor and then through a heat exchanger in which steam is produced. A satisfactory coolant, therefore, must be a fluid that, in addition to having good heat-transfer properties, is stable both to high temperatures and to the action of radiations, and it must have little tendency to capture neutrons. Since the coolant is circulated through the reactor and heat exchanger it should be noncorrosive and nonerosive, and should require relatively low pumping power.

1.28. One of the unusual features of a nuclear reactor is that, in principle, the power output is independent of its size; it is determined essentially by the rate at which heat can be removed from the system. A small reactor could be designed to produce large amounts of power, provided extremely high heat-transfer rates could be achieved. The development of relatively compact reactors with high power density thus depends upon the solution of unusual heat-transfer problems.

1.29. Instead of employing stationary fuel elements and a circulating coolant, reactors have been designed with circulating fuel consisting of a molten alloy of uranium, a mixture of fused salts, or an aqueous solution of a uranium compound. If the fuel is passed directly through a heat exchanger, it could be utilized to produce steam without the intervention of a coolant. This approach is promising, although it involves pumping the intensely radioactive liquid fuel at high temperatures and, possibly, high pressures. In addition, loss of valuable neutrons occurs in the circulating system outside the reactor.

1.30. The use of liquid fuels has attracted interest because it would simplify processing after the fission products have accumulated to such an extent that further operation of the reactor is not possible. A solid fuel element must be dissolved and then refabricated after purification. It was indicated above that the frequent processing of reactor fuel, which is necessary if extensive use is to be made of the available uranium and thorium supplies, will add considerably to the cost of nuclear power. Technological progress in the direction of simplifying fuel processing is thus essential if nuclear power is to be an economic possibility.

Safety Problems in Reactor Design

1.31. The safety aspects of a nuclear reactor system differ from those generally familiar to engineers because of the emission of hazardous radiations, consisting partly of neutrons and partly of beta and gamma radiations from the fission products. The fission product radiations, in particular, persist, although with steadily decreasing intensity, for a long time after the reactor is shut down or the spent fuel elements are discharged. Operating personnel must be protected from the radiations by suitable shielding, which may consist of 6 ft or more of concrete. This shield adds considerably to the size and weight of the reactor, and so represents one of the major difficulties in the design of a mobile nuclear power plant.

1.32. Special methods must be devised for loading the reactor with fresh fuel elements and for their removal after a specified period of operation. The processing of the spent fuel may have to be carried out by remote control, and some way must be found for disposing of the radioactive residues, consisting mainly of fission products for which there is at present little use.

1.33. There is always a possibility that a nuclear reactor might accidentally go out of control. Heat would then be generated by fission faster than it can be removed by the coolant and, as a result, the temperature would rise. In general, reactors are designed so as to have a negative temperature coefficient; that is to say, as the temperature increases they tend to shut themselves down.

Nevertheless, a local rise of temperature might lead to damage to the reactor and possibly to the escape of radioactive material. In order to reduce the chances of this happening, numerous automatic safety devices are included in the reactor design. These must be sensitive enough to shut the reactor down quickly at the first indication of a fault in its operation, but not so sensitive that they prevent the reactor from functioning continuously in spite of inevitable transient fluctuations.

1.34. Liquids and gases which traverse the reactor, either as coolants or for other purposes, are likely to become radioactive. Hence auxiliary equipment, such as pumps, blowers, valves, heat exchangers, etc., must be capable of operating for long periods without maintenance, or if maintenance is essential it must be performed by remote control. Such equipment must be absolutely liquid- and gas-tight, for even small leakages, which would be tolerable in other instances, cannot be permitted.

1.35. In most engineering developments, structural parts or equipment which fail during operation can usually be replaced without difficulty. Because of the radiation hazard, however, this is not true for nuclear reactors. Consequently, much more analytical design work and preliminary testing of various components are necessary in reactor construction, so that the chances of failure after assembly are reduced to a minimum. Further, the design must always be such that if, in spite of all precautions, a failure does occur, it will not have serious consequences.

Problems of Reactor Development

1.36. It should be apparent from the preceding paragraphs why there is much uncertainty, as indicated in § 1.21, concerning the best designs for nuclear reactors. It is because there are still a large number of problems of a technological character, in connection with reactor construction, to which the most satisfactory solutions are not yet known. The necessary information can be obtained only by actually building and operating reactors of various types, and it is the high cost of even the simplest of such reactors that accounts for the relatively slow development of nuclear power.

1.37. There is no general agreement as to the probable cost of a complete electrical power plant utilizing nuclear energy, but estimates range from $200 to $1000 or more per kilowatt of capacity. A full-scale nuclear power plant, with a capacity of 60,000 kw of electricity, sufficient to supply the present requirements of an average American city of nearly 100,000 population, is under construction at Shippingport, Pa. A sum of $85,000,000 has been allotted by the U. S. Atomic Energy Commission for this plant, but much of the expenditure is needed for development work so that the cost per kilowatt is exceptionally high. However, when the plant is in operation, by about 1957, answers should be available to some, but by no means to all, of the unsolved problems of reactor technology.

1.38. The foregoing discussion has referred, in particular, to power reactors since these are probably of greatest interest to engineers, both at present and in the future. The remarks concerning operation, design, and construction apply, nevertheless, to reactors of other types. Several reactors, in the United States and in other countries, are being used for experimental work with neutrons that has no obvious connection with power production. In addition, some nuclear reactors serve to produce radioactive isotopes for various applications in research and industry.

1.39. Reactors have also been built to try out various ideas which might be utilized in nuclear power plants and to test the radiation stability of constructional materials for such plants. Finally, mention may be made of many production reactors whose sole (or main) purpose is to make fissionable material, e.g., plutonium, for weapons, with little or no use being made of the heat generated. The dual-purpose reactors considered by the industrial groups, referred to in § 1.23, would produce plutonium and also utilize the heat for power generation.

Nuclear Reactor Engineering

1.40. The field of endeavor which encompasses the design, construction, and operation of all types of reactors and their associated auxiliaries is referred to as nuclear reactor engineering. It should be emphasized that reactor engineering is based upon well-established aspects of engineering, although their application is somewhat novel in character and is restricted by the limitations arising from the nature of the fission process. In order that trained engineers may be able to apply their knowledge and experience to solving the problems outlined above, it is necessary that they understand the basic principles underlying the processes and reactions which make possible the release of nuclear energy. It is the main purpose of this book to elucidate these principles.

1.41. In the remainder of this introductory chapter, the broad fundamentals of reactor design and operation will be reviewed. It is hoped thereby to present a general over-all picture of the many factors which must be taken into consideration in the construction of a nuclear reactor. It will be apparent that the complete design of a reactor represents a cooperative effort in which biologists, chemists, mathematicians, and physicists work with engineers. Hence, it is desirable that the nuclear reactor engineer be familiar with those aspects of the various branches of science having a direct bearing upon the solution of reactor problems.

ATOMIC STRUCTURE

Atomic Number and Mass Number

1.42. The operation of a nuclear reactor depends upon various interactions of neutrons with atomic nuclei. In order to understand the nature and charac-

teristics of these reactions it is desirable to consider briefly some of the fundamentals of atomic and nuclear physics.*

1.43. An atom consists of a positively charged nucleus surrounded by a number of negatively charged particles, called *electrons*, so that the atom as a whole is electrically neutral. Atomic nuclei are built up of two kinds of primary particles, namely, *protons* and *neutrons*. The proton carries a single unit positive charge, equal in magnitude to the electronic charge. It is, in fact, identical with the nucleus of a hydrogen atom, i.e., a hydrogen atom minus its single electron. The mass of the proton is 1.00758 atomic mass units (amu).† The neutron is an electrically neutral particle carrying no charge; its mass is 1.00897 amu.

1.44. For a given element, the number of protons present in the atomic nucleus, which is the same as the number of positive charges it carries, is called the *atomic number* of the element and is usually represented by the symbol Z. It is identical with the ordinal number of the element in the familiar periodic table of the elements. Thus the atomic number of hydrogen is 1, of helium 2, of lithium 3, and so on, up to 92 for uranium, the element of highest atomic weight existing in nature to any appreciable extent. A number of heavier elements have been made artificially; of these, plutonium, atomic number 94, is important in connection with the release of nuclear energy.

1.45. The total number of protons and neutrons in an atomic nucleus is called the *mass number* of the element and is denoted by A. The number of protons is Z, as stated above, and so the number of neutrons in the atomic nucleus is $A - Z$. Since the masses of both neutron and proton are close to unity, on the atomic mass scale, it is evident that the mass number is the integer nearest to the atomic weight of the species under consideration.

Isotopes

1.46. It is the atomic number, i.e., the number of protons in the nucleus, which determines the chemical nature of an element. This is so because the chemical properties depend on the (orbital) electrons surrounding the nucleus, and their number must be equal to the number of protons, since the atom as a whole is electrically neutral. Consequently atoms with nuclei containing the same numbers of protons, i.e., with the same atomic number, but with different mass numbers, are essentially identical chemically, although they frequently exhibit marked differences in their nuclear characteristics. Such species, having the same atomic number but different mass numbers, are called *isotopes*. They are, in general, indistinguishable chemically, but have different atomic weights.

1.47. The element uranium, which is at present the most important for the release of nuclear energy, exists in nature in at least three isotopic forms, with mass numbers 234, 235, and 238, respectively. It is usual to distinguish among

* See also, S. Glasstone, "Sourcebook on Atomic Energy," D. Van Nostrand Co., Inc., 1950.

† An atomic mass unit is 1.660×10^{-24} gram. The actual mass of any atom is equal to its (physical) atomic weight in mass units.

the different isotopes of an element by writing the mass number after the name or symbol; thus, the heaviest isotope of uranium would be represented as uranium-238, U-238, or U^{238}. The proportions in which the isotopes occur in natural uranium and the weights of the respective atoms are given in Table 1.1. It is seen that uranium-238 is by far the most abundant isotope, but all natural uranium contains a little over 0.7 per cent of uranium-235. Both of these isotopes play significant parts in the field of nuclear energy. The proportion of uranium-234 is so small that it can be ignored.

TABLE 1.1. ISOTOPIC COMPOSITION OF NATURAL URANIUM

Mass Number	*Per Cent*	*Atomic Mass* (amu)
234	0.006	234.11
235	0.712	235.12
238	99.282	238.12

1.48. Another element which may ultimately prove to be important from the nuclear energy standpoint is thorium, atomic number 90. This occurs in nature almost entirely as a single nuclear species, with mass number 232. There are traces of other isotopic forms, but their proportions are negligible.

INTERACTIONS OF NEUTRONS WITH NUCLEI

Scattering Processes

1.49. The release of nuclear energy in a form capable of practical utilization results from a particular type of interaction of neutrons with a very few nuclear species, one of which is uranium-235. Neutrons can be obtained in the free state, i.e., outside atomic nuclei, in various ways (§ 2.71). Because they are uncharged, they suffer no electrical repulsion, as do positively charged particles, when they approach an atomic (positively charged) nucleus. Consequently, whereas charged particles, e.g., protons, must have large amounts of kinetic energy to overcome the repulsive electrical forces before they can interact with atomic nuclei, neutrons can take part in nuclear reactions when they have very little kinetic energy.

1.50. When produced in the free state, by ejection from atomic nuclei, neutrons almost invariably have high energies, usually of the order of 1 to 10 million electron volts (Mev).* Since the mass of a neutron is roughly 1 amu, i.e., about 1.7×10^{-24} gram, it can be readily calculated that these high-energy neutrons move with velocities of the order of 10^9 cm/sec, i.e., about one tenth of the velocity of light. Such neutrons are consequently called *fast neutrons*.

* In atomic studies, energies are frequently expressed in *electron volt* units. The electron volt, i.e., 1 ev, is the energy acquired by any charged particle carrying a unit charge when it passes, without resistance, through a potential difference of 1 volt. It is equivalent to 1.60×10^{-12} erg. The million electron volt (or Mev) unit is 10^6 ev or 1.60×10^{-6} erg.

1.51. In their passage through matter neutrons collide with atomic nuclei and scattering occurs in which there is a transfer of energy from the fast neutrons to the relatively slow-moving nuclei. These scattering collisions are of two types, namely, inelastic and elastic. In an *inelastic scattering* process momentum is conserved, but kinetic energy is not. Part of the kinetic energy of the neutron is converted into internal (potential) energy, sometimes referred to as *excitation energy*, of the struck nucleus. Inelastic neutron scattering is related to specific properties of the scattering nucleus and, in general, occurs only when the kinetic energy of the neutron is fairly large. For elements of moderate or high mass number, the neutron must possess about 0.1 Mev of energy, at least, if it is to be involved in an inelastic collision process. If the scattering material has a low mass number, the required neutron energy is even higher.

1.52. In *elastic scattering* there is strict conservation of both momentum and energy. Some (or all) of the kinetic energy of the neutron appears, after the collision, as kinetic energy of the struck nucleus. The process may be treated by the laws of classical mechanics, as a "billiard ball" type of collision. In each elastic collision with an essentially stationary nucleus, the neutron will transfer part (or all) of its kinetic energy to the nucleus and is thereby slowed down. For a nucleus of given mass the amount of energy transferred will depend on the angle through which the neutron is scattered. For a given scattering angle the fraction of the neutron energy transferred and, hence, the extent of slowing down will be greater the smaller the mass of the scattering nucleus.

Thermal Neutrons

1.53. Below energies of about 0.1 Mev, inelastic neutron collisions do not occur, but elastic collisions between neutrons and nuclei will be effective in slowing down the neutrons until their average kinetic energy is the same as that of the atoms (or molecules) of the scattering medium. This energy depends on the temperature of the medium, and hence it is called *thermal energy*. Neutrons whose energies have been reduced to values in this region are designated *thermal neutrons*. The average energy of a thermal neutron is given by

$$\begin{aligned}\text{Average energy of thermal neutrons} &= 8.6 \times 10^{-5}\, T_K \text{ ev} \\ &= 4.8 \times 10^{-5}\, T_R \text{ ev},\end{aligned}$$

where T_K and T_R are the absolute temperatures of the scattering medium on the Kelvin and Rankine scales, respectively. Thus, at ordinary temperatures, i.e., about 22°C or 295°K (72°F or 532°R), the average energy of a thermal neutron is approximately 0.025 ev.

1.54. The average speed of a thermal neutron is expressed by

$$\begin{aligned}\text{Average speed of thermal neutron} &= 1.3 \times 10^4 \sqrt{T_K} \text{ cm/sec} \\ &= 0.97 \times 10^4 \sqrt{T_R} \text{ cm/sec},\end{aligned}$$

so that at ordinary temperatures it is about 2.2×10^5 cm/sec. The average energies and speeds of thermal neutrons at a number of temperatures are given in Table 1.2.

TABLE 1.2. AVERAGE SPEEDS AND ENERGIES OF THERMAL NEUTRONS

Temperature		*Energy*	*Speed*
(°C)	(°F)	(ev)	(cm/sec)
25	77	0.026	2.2×10^5
200	392	0.041	2.8
400	752	0.058	3.4
600	1112	0.075	3.8
800	1472	0.092	4.2

1.55. In nuclear reactor work, the term *fast neutrons* is applied to neutrons having energies of about 0.1 Mev, i.e., 10^5 ev, or more. Those with energies from 10^5 ev down to 1 ev are called *intermediate neutrons*, whereas those having energies below about 1 ev are referred to as *slow neutrons*. Thermal neutrons are slow neutrons which are in thermal (or molecular kinetic) equilibrium with their surroundings. Neutrons with energies just above the thermal region, i.e., in the low intermediate range, are sometimes named *epithermal neutrons*.

1.56. The slowing down of neutrons plays an important part in many nuclear reactors, and the material used for the purpose is called a *moderator*. The process of slowing down as a result of scattering collisions is referred to as *moderation*. A good moderator is a material which reduces the speed of fast neutrons in a small number of collisions. Consequently, materials consisting of atoms of low mass number (§ 1.45) are the best moderators. Ordinary water (H_2O), heavy water (D_2O), beryllium, beryllium oxide, and carbon have been employed as moderators in various reactors. The fact that helium is a gas makes its use as a moderator impractical in a nuclear reactor. A good moderator must not absorb neutrons to any extent; for this reason the light elements lithium and boron are ruled out.

Radiative Capture Reactions

1.57. Neutrons can take part in several different types of capture reactions with atomic nuclei, but only two, which are important in nuclear reactors, will be considered here. In one type of reaction, called *radiative capture*, the nucleus captures the neutron and forms what is called a *compound nucleus* in a high-energy (or excited) state. The excess energy is then emitted in the form of electromagnetic radiation, referred to as *capture gamma rays*, so that the compound nucleus is left in its normal (or ground) state. The process may be represented symbolically by the equation

$$ {}_zX^A + {}_0n^1 \rightarrow [{}_zY^{A+1}]^* \rightarrow {}_zY^{A+1} + \gamma,$$

where X is the nucleus undergoing reaction, n represents the captured neutron, and Y is the compound nucleus formed as a result of the capture. The subscripts indicate the number of positive charges, or protons, i.e., the atomic number Z for the nucleus X and zero for the neutron. The superscripts give the number of neutrons plus protons, i.e., the mass number A for the nucleus X and unity for the neutron. In any reaction, the sum of the subscripts as well as that of the superscripts must remain unchanged. The excited (high-energy) compound nucleus is marked by an asterisk, and γ represents the gamma radiation emitted.

1.58. The product Y of the radiative capture reaction is seen to be an isotope of X, since it has the same atomic number Z, but with a mass number one unit greater. In many cases, the Y nucleus is somewhat unstable and exhibits the phenomenon of *radioactivity*. It undergoes spontaneous change, at a definite exponential rate (§ 2.5), into another type of nucleus, at the same time emitting certain characteristic radiations. In the case under consideration the nuclear radiations almost invariably consist of electrons, referred to in this connection as negative *beta particles*,* frequently accompanied by gamma rays which carry off excess energy. The beta particle bears a single negative charge and has a mass of about 0.00055 amu; its mass number is thus taken as zero. The product of radioactive beta decay is then a nucleus with the same mass number as its parent, but with a positive charge (or atomic number) one unit greater, as will be seen shortly. In some instances the decay product is itself radioactive and undergoes a further stage of beta decay, as described below.

1.59. Radiative capture reactions are designated by the symbol (n, γ), implying that a neutron is captured and a gamma ray is emitted in the process. Several such reactions occur in nuclear reactors, and two of special interest will be mentioned here. The first is the capture of neutrons by uranium-238, the most abundant naturally occurring isotope of this element (see Table 1.1); the (n, γ) process may be represented as

$$_{92}U^{238} + {}_{0}n^{1} \rightarrow {}_{92}U^{239} + \gamma.$$

The resulting nucleus, uranium-239, is radioactive and decays with the emission of a negative beta particle, indicated by $_{-1}\beta^{0}$ (charge -1, mass number zero); thus,

$$_{92}U^{239} \rightarrow {}_{-1}\beta^{0} + {}_{93}Np^{239},$$

the product being an isotope of an element of atomic number 93, called *neptunium*, Np, which does not exist in nature to any detectable extent. Neptunium-239 is also beta-active and decays fairly rapidly, according to the process

$$_{93}Np^{239} \rightarrow {}_{-1}\beta^{0} + {}_{94}Pu^{239},$$

to form the isotope Pu^{239} of the element of atomic number 94, called *plutonium*.

1.60. The element plutonium occurs in nature in the merest traces only. Nevertheless, plutonium-239, which may be regarded as an isotope of an artificial element, is being made in appreciable quantities in the nuclear reactors at Hanford, Washington, and will also be produced at the Savannah River plant, near Aiken, South Carolina. It is obtained from uranium-238, as a result of radiative capture of neutrons, released in reactors, followed by two relatively rapid stages of beta decay, as stated above. Plutonium-239 is also radioactive, although it decays quite slowly; it expels *alpha particles*, which are, in fact, identical with helium nuclei, since they have a mass of four units and carry a double positive charge. The importance of plutonium-239 lies in the fact that it can be used for the release of nuclear energy both in atomic bombs and in nuclear reactors.

1.61. A series of processes similar to that just described is initiated by the (n, γ) reaction with the naturally occurring thorium-232; thus,

$$_{90}Th^{232} + {}_{0}n^{1} \rightarrow {}_{90}Th^{233} + \gamma,$$

so that the product is the isotope thorium-233. This undergoes two successive stages of beta decay, the first being

$$_{90}Th^{233} \rightarrow {}_{-1}\beta^{0} + {}_{91}Pa^{233},$$

where Pa^{233} is the symbol for protactinium-233, and the second

$$_{91}Pa^{233} \rightarrow {}_{-1}\beta^{0} + {}_{92}U^{233},$$

so that the product is uranium-233, an isotope of this element which does not occur in any appreciable amount in nature. Like plutonium-239, it emits alpha particles at a relatively slow rate, and, although it is not yet being made in quantity, it is likely to play a significant part in the release of nuclear energy.

1.62. The radiative capture process has been illustrated here by reference to two elements of high atomic number, but essentially all the elements, from hydrogen to uranium, undergo the reaction to a greater or lesser extent. Thus, with hydrogen the process is

$$_{1}H^{1} + {}_{0}n^{1} \rightarrow {}_{1}H^{2} + \gamma;$$

the product, represented here by the symbol H^2, is the stable, heavier isotope of hydrogen, i.e., deuterium, frequently called "heavy hydrogen." Although this reaction is not used to make deuterium, it is significant in certain aspects of nuclear reactor design.

* The emission of electrons from a radioactive nucleus does not mean that these particles are actually present as such in the nucleus. They are believed to arise from the transformation within the nucleus of a (neutral) neutron into a (positive) proton plus a (negative) electron; the proton remains in the nucleus and the electron is expelled as beta radiation. It is probable that at the same time another (neutral) particle of very small mass, called a *neutrino*, is produced which carries off a large proportion of the energy accompanying the beta-decay process.

Rates of Nuclear Reactions

1.63. The rates of nuclear reactions are conveniently expressed by making use of a quantity called the *nuclear cross section*, represented by the symbol σ. This quantity is of great importance and will be considered more fully in Chapter II, but for the present it is sufficient to say that the cross section for a reaction is a measure of the probability of the occurrence of that reaction under prescribed conditions. It is a specific property of the nucleus for a particular reaction with incident particles, e.g., neutrons, of a given energy.

1.64. Determinations of the cross sections for radiative capture reactions reveal some interesting facts. Although the actual capture cross sections for neutrons of a given energy vary greatly from one nuclear species to another, they generally show a similar trend as the neutron energy is changed. The capture cross sections are relatively high for neutrons of low energy, i.e., for slow neutrons, and are appreciably lower for neutrons of high energy, i.e., for fast neutrons. Because many slow-neutron reactions with atomic nuclei (not only radiative capture) occur more rapidly than do those with fast neutrons, the latter are often slowed down deliberately, in order to increase the probability of interaction. This is the purpose of the moderating process, referred to above, used in many nuclear reactors.

1.65. In the slow-neutron region, the capture cross section decreases with increasing neutron velocity in such a manner that the so-called "$1/v$ law" is obeyed. That is to say, in this region, the cross section is inversely proportional to the velocity, v, of the neutron relative to that of the nucleus. Physically, this may be thought of as implying that the probability of interaction between a given nucleus and a neutron is determined by the time, which is proportional to $1/v$, spent by the neutron in the vicinity of the nucleus.

1.66. In the low intermediate range, e.g., for neutrons of about 1 to 1000 ev energy, especially for elements of moderate and high mass number, there are often specific energies for which the rate of a given nuclear reaction, and hence the cross section, is exceptionally large. The phenomenon is attributed to what is called *resonance* (§ 2.132). Neutrons having energies equal (or close) to the resonance value (or values) have a much greater probability of undergoing the given reaction than do those with somewhat higher or lower energies. Uranium-238, for example, exhibits three marked resonances for neutrons with energies of 6.5, 21, and 36 ev, respectively, as well as a number of others (see Fig. 2.18).

THE RELEASE OF NUCLEAR ENERGY

The Nuclear Fission Process

1.67. The second type of neutron reaction of immediate interest is *nuclear fission*, or, in brief, fission. In the fission process the nucleus absorbs a neutron and the resulting compound nucleus is so unstable that it immediately breaks up into two parts of more or less equal mass, called *fission fragments*. There

are many different ways in which fission of a particular nucleus takes place, although in only a small fraction of the fissions does the nucleus break up in a symmetrical manner. Consequently, there are many different fission fragments formed when a particular nucleus undergoes fission. Most of the fragments are radioactive, decaying at different rates, with the emission of negative beta particles and gamma radiation, to form products which are themselves usually radioactive. In the fission of uranium-235, for example, there are formed more than 80 primary products, with mass numbers ranging from 72 to 160. Each of these undergoes, on the average, three stages of radioactive decay before being converted into a stable nucleus. As a result, there are over 200 radioactive isotopes of 30 or more different elements present among the fission products after a short time.

1.68. Nuclear fission as a result of neutron capture occurs only with the heaviest elements, those of special interest being thorium, uranium, and plutonium. Whereas certain isotopes, notably uranium-233, uranium-235, and plutonium-239, undergo fission with slow, e.g., thermal, neutrons, as well as with fast neutrons, others, such as thorium-232 and uranium-238, require fast neutrons to cause fission. Where fission is possible by slow neutrons, the variation of fission cross section with neutron energy is similar to that described in § 1.64. In general, the cross section is largest for thermal neutrons; it then decreases with increasing neutron energy, according to the $1/v$ law, and becomes relatively small for fast neutrons.

1.69. For thorium-232 and uranium-238, there is a fairly sharp energy threshold at about 1 Mev; neutrons with energy less than this amount are unable to cause fission to any appreciable extent. Nevertheless, it is of interest to note that both of these species, and especially the latter, undergo spontaneous fission. Thus, in 1 gram of uranium-238 about 24 nuclei suffer spontaneous fission per hour. This fact is of significance in connection with the start-up of certain reactors.

1.70. There are good theoretical reasons for believing that nuclei capable of undergoing fission by slow neutrons, but which are otherwise reasonably stable, are those of high mass number having odd numbers of neutrons. The only nuclear species existing in nature to any appreciable extent which satisfies these requirements is uranium-235. Since its mass number, i.e., number of neutrons plus protons, is 235, and its atomic number, i.e., number of protons, is 92, it has an odd number of neutrons, i.e., $235 - 92 = 143$, in its nucleus. The artificially prepared uranium-233 and plutonium-239 also have odd numbers of neutrons and are fissionable by slow neutrons. It seems unlikely that any other, relatively stable, substances having this property will be available in sufficient quantities to be of practical interest.

1.71. From the point of view of the utilization of nuclear energy, the importance of fission lies in two facts. First, the process is associated with the release of considerable amounts of energy, and second, the reaction initiated by neutrons

is also accompanied by the liberation of neutrons. It is thus possible, under proper conditions, for the process to be self-sustaining and for energy to be generated continuously, once the fission reaction has been started. These two aspects of fission, which are fundamental to the operation of nuclear reactors, will be considered in reverse order.

Release of Neutrons in Fission

1.72. Most of the nuclear fragments formed when fission occurs have too many neutrons for stability, and so there will be a tendency for some of them to expel neutrons almost instantaneously. These are the neutrons which appear to accompany the fission process. The average number, ν, of neutrons liberated for each thermal neutron absorbed in a fission reaction by uranium-235 and plutonium-239 is given in Table 1.3. It will be noted that the numbers of neutrons liberated are not integers. This is because the nucleus splits in many different ways, as already mentioned, and, although the number of neutrons expelled in any individual act of fission must be integral, the average is not necessarily a whole number.

TABLE 1.3. AVERAGE NUMBER OF NEUTRONS LIBERATED PER THERMAL NEUTRON ABSORBED IN FISSION

Fissionable Nucleus	*Number of Neutrons* (ν)
Uranium-235	2.5 ± 0.1
Plutonium-239	3.0 ± 0.1

1.73. The neutrons emitted as a result of the fission process can be divided into two categories, namely, *prompt neutrons* and *delayed neutrons*. The prompt neutrons, which constitute over 99 per cent of the fission neutrons, are released within an extremely short interval of time, probably about 10^{-14} sec (or less), of the instant of fission. The energies of these neutrons cover a considerable range, from over 10 Mev down to quite small values; the majority, however, have energies of about 1 to 2 Mev (§ 2.166).

1.74. Whereas the expulsion of the prompt neutrons ceases immediately after fission, the delayed neutrons, as their name implies, are emitted over a period of a few hours, the intensity falling off rapidly with time. The delayed neutrons accompanying fission fall into five major groups. The rate of decay of the intensity in each delayed-neutron group is exponential, as it is for radioactive change. The same five groups of delayed neutrons accompany the fission of uranium-233, uranium-235, and plutonium-239, although the total fraction of delayed neutrons and their distribution among the groups are different in each case. It is of special interest to note that the properties of the delayed neutrons have an important bearing on the time-dependent behavior of nuclear reactors. Were it not for these neutrons, the safe control of nuclear reactors would be much more difficult than it is.

Energy of Fission

1.75. The fission process is remarkable for the magnitude of the energy released; it is about 200 Mev for each nucleus undergoing fission, which may be compared with a few electron volts for each atom reacting in chemical processes, such as the combustion of coal or oil. The large energy release in fission is associated with the fact that the products of the reaction have an appreciably smaller total mass than that of the nucleus undergoing fission plus the neutron causing fission. Because of the equivalence of mass and energy, which has now been firmly established from the study of many nuclear processes, the considerable decrease of mass in the fission reaction must be accompanied by the liberation of a large amount of energy.

1.76. The relationship between the energy E and the equivalent mass m is given by the Einstein equation

$$E = mc^2, \tag{1.76.1}$$

where c is the velocity of light. If m is in grams, and c in cm/sec, i.e., 3×10^{10} cm/sec, then E will be in ergs. For the present purpose it is more useful to express m in atomic mass units (§ 1.43); equation (1.76.1) then becomes

$$E(\text{ergs}) = m(\text{amu}) \times 1.49 \times 10^{-3}.$$

As seen in § 1.50, 1 Mev $= 1.60 \times 10^{-6}$ erg, and so the energy equivalent is expressed in Mev by

$$E(\text{Mev}) = m(\text{amu}) \times 931. \tag{1.76.2}$$

In other words, 1 amu is equivalent to 931 Mev.

1.77. The magnitude of the energy released in nuclear fission will be estimated for the fission of uranium-235, making the simplifying assumption that the products are nuclei with mass numbers 95 and 139, since these are known to be obtained in greatest amount. In order to balance the mass numbers, it is evident that two fission neutrons are liberated in this case, as may be seen from the equation

$$U^{235} + n^1 \rightarrow X^{95} + Y^{139} + 2n^1,$$

the neutron on the left side being the one which initiates the fission, and the two on the right side those which are formed as a result of fission.

1.78. The mass of the uranium-235 atom is known to be 235.124 amu, whereas that of the neutron is 1.00897 amu, which may be approximated to 1.009 for the present purpose. By comparison with known stable species, the masses of the two fission products postulated above will be 94.945 and 138.955 amu, respectively.* Hence the following balance sheet of the masses before and after fission may be drawn up:

* It should be noted that the masses used here are not those of the nuclei, but of the respective atoms, which are determined by experiment. The values include the masses of the outer (or orbital) electrons. In calculating the decrease of mass in the fission process, the masses of the electrons cancel, since they are the same before and after the reaction.

Masses Before Fission		*Masses After Fission*	
Uranium-235	235.124	Mass number-95	94.945
1 neutron	1.009	Mass number-139	138.955
		2 neutrons	2.018
Total	236.133	Total	235.918

$$\text{Mass converted into energy} = 236.133 - 235.918 = 0.215 \text{ amu.}$$

As seen above, 1 amu is equivalent to 931 Mev, and so the energy released per fission is given by

$$\text{Energy released per fission} = (0.215)(931) = 198 \text{ Mev.}$$

1.79. Although this calculation was made for one particular mode of fission, it may be regarded as quite typical. While there are slight variations from one mode to another, it appears, on the whole, that an estimate of about 200 Mev of energy released per uranium-235 nucleus undergoing fission is satisfactory. The same value may also be taken as applying to the fission of uranium-233 and plutonium-239.

Distribution of Fission Energy

1.80. The major proportion — over 80 per cent — of the energy of fission appears as kinetic energy of the fission fragments, and this immediately manifests itself as heat. Part of the remaining 20 per cent or so is liberated in the form of instantaneous gamma rays and as kinetic energy of the fission neutrons.* The rest is released gradually as energy carried by the beta particles and gamma rays emitted by the radioactive fission products as they decay over a period of time.

1.81. The approximate distribution of the fission energy, which may be regarded as applying to all three of the important fissionable species, is given in Table 1.4. The energy represented by the items in the first three lines is released

TABLE 1.4. DISTRIBUTION OF FISSION ENERGY

	Mev
Kinetic energy of fission fragments	168 ± 5
Instantaneous gamma-ray energy	5 ± 1
Kinetic energy of fission neutrons	5 ± 0.5
Beta particles from fission products	7 ± 1
Gamma rays from fission products	6 ± 1
Neutrinos	~ 10
Total fission energy	201 ± 6

* This may be regarded as including the energy of the delayed neutrons, which constitutes probably less than 0.02 per cent of the total fission energy.

at the time of fission, whereas that in the next two lines is set free gradually as the fission products decay. The (10 Mev) energy of the neutrinos, which accompany the beta radioactivity, is not available for producing power since these particles do not interact appreciably with matter.

1.82. The values in the table give what may be called the primary fission energy, of which about 191 Mev is available per fission.* There is a further, or secondary, source of energy which arises when fission occurs in a nuclear reactor. Some of the excess fission neutrons are captured by the nuclei of various materials, e.g., uranium-238, moderator, coolant, structure, etc., present in the reactor, to undergo radiative capture, i.e., (n, γ), reactions. Thus neutrons are absorbed and energy is released in the form of gamma radiation. Since neutron capture and emission of gamma radiation occur within a very short time, this energy may be regarded as being released instantaneously, i.e., at the moment of fission. The energy of the capture gamma radiation will depend on the materials present in the reactor, but it may be taken to be 3 to 7 Mev/fission. Since the products of the (n, γ) reaction are frequently radioactive, perhaps another 1 or 2 Mev, on the average, may be released over a period of time, as beta-particle and gamma-ray energy, while these products decay. The secondary energy resulting from radiative capture of some of the fission neutrons is thus approximately equal to the energy carried off by neutrinos. The total energy available in a reactor is therefore roughly 195 to 200 Mev/fission; the latter figure will be used in the subsequent discussion.

1.83. All the energy released within the reactor, disregarding the neutrino energy, is ultimately transformed into heat; the various mechanisms whereby this takes place are as follows:

(1) Transfer of kinetic energy from fission products.
(2) Transfer of kinetic energy from fission neutrons.
(3) Deceleration and absorption of beta particles.
(4) Absorption of gamma rays.

The conversion of the beta-particle and gamma-ray energies into heat actually involves several stages (see Chapter II), but the summarizing statements above are sufficient for the present. Since the fission products and the beta particles which they emit do not travel very far, the heat they produce may be regarded as being released essentially at the point of fission. As the fission neutrons and gamma radiations of various kinds, e.g., instantaneous fission gamma rays, capture gamma rays, and those accompanying radioactive decay, can travel considerable distances before absorption, their energy may be liberated at some distance from the place where fission occurs.

1.84. Some of the foregoing results are reviewed in Table 1.5, in which is given an indication of the heat produced almost instantaneously and that liberated

* Measurements in a reactor gave a value of 190 ± 5 Mev/fission; see J. L. Meem, *Nucleonics*, **12**, No. 5, 62 (1954).

TABLE 1.5. LIBERATION OF HEAT DUE TO FISSION

Instantaneous	Mev
Energy of fission fragments	168
Energy of fission neutrons	5
Instantaneous gamma rays	5
Capture gamma rays	7
	185
Delayed	
Beta particles from fission products	7
Gamma rays from fission products	6
Radiations from capture products	2
	15

gradually over a period of time. Of the heat liberated instantaneously, about 90 per cent, i.e., 168 Mev, will be produced at (or near) the point of fission, and only 17 Mev elsewhere. But of the delayed heat, the two amounts are approximately equal, i.e., about 7 Mev at the place where fission occurs and 8 Mev at a distance.

1.85. There are several interesting conclusions to be drawn from these data in connection with the operation of nuclear reactors. In the first place, when the reactor is started up, only 185 Mev of heat will be released per fission. In the course of time, as the fission products and capture products accumulate and decay, the heat will increase gradually to 200 Mev, at which value it will remain constant, provided the system is in equilibrium. When the reactor is shut down, that is to say, when the fission process is brought to a virtual stop, the delayed heat will continue to be released for some time while the various radioactive nuclei decay. Only a fraction of this heat — perhaps no more than half — is liberated in the fissionable material, the remainder being produced elsewhere, e.g., in the moderator, structure, etc. These matters have an important bearing on the problems of heat removal from nuclear reactors.

Thermal Values of Fissionable Material

1.86. In order to convert the fission energy values into practical units, it should be recalled that 1 Mev is equal to 1.60×10^{-6} erg, and this is equivalent to 1.60×10^{-13} watt-sec. Hence the total energy (200 Mev) available per fission is about 3.2×10^{-11} watt-sec, so that it requires 3.1×10^{10} fissions to release 1 watt-sec of energy. In other words, fissions at the rate of 3.1×10^{10} per sec produce 1 watt of power, provided the reactor has been operating for some time.

1.87. One gram atom, i.e., the atomic weight expressed in grams, of any element contains the Avogadro number, i.e., 6.02×10^{23}, of individual nuclei, and if all of these undergo fission, the energy liberated would be $(6.02 \times 10^{23})(3.2 \times 10^{-11}) = 1.9 \times 10^{13}$ watt-sec, i.e., 5.3×10^{6} kw-hr. This is the amount of heat that would be released by the complete fission of 233 grams of uranium-233, or 235 grams of uranium-235, or 239 grams of plutonium-239. Neglecting

the relatively small differences between these weights, the results in Table 1.6 may be regarded as applying to the heat produced by the fission of one pound of any of these materials.* A useful fact to remember is that the power production corresponding to the fission of 1 gram of material per day would be roughly 10^6 watts, i.e., 1 megawatt.

TABLE 1.6. HEAT LIBERATED BY 1 LB OF FISSIONABLE MATERIAL

0.9×10^{13} cal
1.0×10^{7} kw-hr
2.8×10^{13} ft-lb
3.6×10^{10} Btu

THE FISSION CHAIN IN NUCLEAR REACTORS

Conditions for Self-sustaining Chain Reaction

1.88. Since neutrons are produced in the fission reaction initiated by neutrons, this process obviously has the potentialities of a chain reaction. But for application in a nuclear reactor, the chain must be self-sustaining and must not die out after a relatively small number of stages. If every neutron liberated when fission occurs were able to cause fission of another nucleus, there would be no doubt that the chain could be maintained, since more than two neutrons are released, on the average, in each act of fission. It is important to bear in mind, however, that, for various reasons, a fraction only of the fission neutrons is available, in actual practice, for the initiation of further fission.

1.89. In the first place, there may not be enough neutrons of sufficient energy to maintain the chain. This is the case, for example, for pure uranium-238 or thorium-232, which require neutrons of at least 1-Mev energy to cause fission. It is true that a large proportion of the fission neutrons do have energy in excess of this amount, but as a result of noncapture collisions — particularly inelastic collisions — with atomic nuclei, the energies are quickly reduced below the fission threshold values. A self-sustaining chain reaction due to fission of uranium-238 or thorium-232 is thus not possible. This situation does not arise with uranium-233, uranium-235, or plutonium-239, since there is no lower limit to the neutron energy for fission to occur.

1.90. Second, it should be noted that neutrons produced by fission can take part in various nonfission reactions. Even in pure fissionable material, not all the neutrons absorbed will cause fission. This is because there is a definite probability, depending on the neutron energy, that other processes, especially radiative capture, will occur. For example, of the thermal neutrons absorbed in pure uranium-235, about 84 per cent cause fission; in plutonium-239, the proportion is only 65 per cent (see § 1.105). In addition, various nonfissionable nuclei

* Because the fissionable materials undergo other reactions, the heat liberated per pound *consumed* is somewhat less (cf. § 1.90).

present will capture some of the neutrons, which will thus not be available for causing fission. Finally, because of the finite size of the system, neutrons will inevitably escape from the surface and will be lost to the chain reaction.

1.91. A nuclear reactor may be regarded as consisting of the fuel * containing the fissionable material, e.g., uranium-233, uranium-235, or plutonium-239, possibly a moderator to slow down the neutrons, a coolant to remove the heat generated by fission, and structural material. The practicability of a self-sustaining chain reaction depends on the relative extents to which the neutrons take part in four main processes:

(1) Complete loss of neutrons from the system, generally referred to as *leakage*.

(2) Nonfission capture by the fissionable nuclei, e.g., uranium-235, and by other nuclei, e.g., uranium-238 or thorium-232, present in the fuel. This is sometimes designated *resonance capture*, since it is most likely to occur at resonance energies (§ 1.66).

(3) Nonfission capture, called *parasitic capture*, by the moderator and various other substances ("poisons"), such as coolant, structure, fission products, and impurities in the fuel and moderator.

(4) Fission capture of neutrons of various energies by the fissionable material, and of fast neutrons by uranium-238 or thorium-232, if present.

1.92. In each of the four processes summarized above neutrons are removed from the system, but in the fourth process, i.e., in the fission reaction, other neutrons are generated to replace them. If the number of neutrons produced in the last process is just equal to (or exceeds) the total number lost by escape and by fission and nonfission capture, a self-sustaining chain reaction should be possible. An illustration of the type of neutron balance that might exist in a reactor using natural uranium as fuel is depicted below. It is assumed that fission results only from the capture of slow neutrons, and that, on the average, 2.5 fast neutrons are produced in each act of fission.

100 fast (fission) neutrons
| ⟶ 5 leak out during slowing down
↓ ⟶ 10 resonance absorptions by U-238 during slowing down
85 slow neutrons produced
↓ ⟶ 5 leak out as slow neutrons
80 slow neutrons available for absorption
| ⟶ 33 absorbed by U-238, moderator, poisons, etc.
↓ ⟶ 7 nonfission capture by U-235
40 absorbed by U-235 to cause fission
↓
100 fast (fission) neutrons.

* The general term "fuel" is used here for the reactor core material in which the fission chain is maintained. It may consist of (essentially) pure fissionable isotopes or nonfissionable species may be present, e.g., uranium-238 when natural uranium (or uranium partially enriched in uranium-235) is the fuel.

1.93. It is seen from this hypothetical case, which might apply to a reactor in which natural uranium is the fuel, that a total of 100 neutrons is either absorbed or leaks out, and that 100 neutrons are produced by fission. This condition represents a stationary, self-sustaining chain reaction. If a total of more than 60 neutrons had leaked out and been captured in nonfission processes, there would have been less than 40 slow neutrons available to cause fission of uranium-235, and consequently less than 100 fast fission neutrons would have been produced. Such a chain would be convergent, and the reaction would virtually cease in a short time. On the other hand, if the leakage and nonfission capture accounted for less than 60 neutrons, there would be more than 40 fissions, and more than 100 fast fission neutrons would result. Such a chain would be divergent, and the number of neutrons present in the system would increase steadily.

The Multiplication Factor

1.94. The conditions for a chain reaction can be conveniently expressed in terms of a useful quantity called the *effective multiplication factor*, k_{eff}. It is defined as the ratio of the average number of neutrons produced by fission in each generation to the total number of corresponding neutrons absorbed by fuel, moderator, etc., or leaking out, on the average. If $k_{\text{eff}} = 1$, i.e., the number of neutrons produced is equal to the total number lost by absorption and leakage, the chain is stationary and is self-sustaining; such a system is said to be *critical*. If $k_{\text{eff}} < 1$, the chain is convergent and is not self-sustaining; this is designated as a *subcritical* system. Finally, if $k_{\text{eff}} > 1$, the chain is divergent, and the system is described as *supercritical*.

1.95. The effective multiplication factor can be regarded as being made up of two parts. One is determined by the composition and arrangement of the materials present in the system, whereas the other is dependent upon its size. The former is called the *infinite multiplication factor* and is represented by the symbol k (or k_∞); it is the ratio of the average number of neutrons produced in each generation to the average number of corresponding neutrons absorbed. In other words, it is essentially the value the effective multiplication factor would have if the system were infinitely large and there were no loss of neutrons by leakage. From the definitions of k_{eff} and k, it follows that

$$\frac{k_{\text{eff}}}{k} = \frac{\text{Neutrons absorbed}}{\text{Neutrons absorbed} + \text{neutrons leaking out}} = P,$$

so that the fraction, indicated by P, is a measure of the probability that neutrons will not leak out of the finite system, but will remain until absorbed.* Hence it is possible to write

$$k_{\text{eff}} = kP,$$

where P, called the *nonleakage probability*, is the quantity, referred to above,

* The tacit assumption involved here is that the energy distribution of the neutrons will be the same in a system of finite size as in one that is infinitely large.

which depends on the size of the system. It is obvious, of course, that P will always be less than unity, except for a system of infinite size when it is equal to unity.

Critical Size

1.96. If, for a given composition and arrangement of specified materials, the infinite multiplication factor k exceeds unity, there will be a certain size of the system for which the nonleakage probability P is such that kP, equal to k_{eff}, is just unity. In other words, there will be a particular size of the reactor for which the neutron production, by fission, is exactly balanced by neutron leakage and absorption. This is called the *critical size* and it is that at which the chain reaction will just be self-sustaining.

1.97. Since neutrons are produced throughout the whole of a reactor whereas leakage takes place only from the exterior surface, it is apparent that the nonleakage probability increases as the volume-to-area ratio of the system increases. Consider, for example, a sphere of radius R; the volume-to-area ratio is proportional to R, and so the larger the radius the greater the nonleakage probability, i.e., the closer it approximates to unity. Consequently, if a system is smaller than the critical size, the value of P will be less than for a critical reactor, and kP, i.e., k_{eff}, will be less than unity. It follows, from what has been stated above, that the chain reaction will then be convergent and the system will be subcritical. On the other hand, if the reactor dimensions exceed the critical size for the given composition and arrangement, k_{eff} will be greater than unity and the system will be supercritical, capable of supporting a divergent fission chain.

1.98. One of the quantities which determines the critical size is evidently the infinite multiplication factor. If this is much larger than unity, then P can be relatively small, and the requirement for a critical system, i.e., $kP = 1$, can still be met. In other words, if k is large, then the critical size will be fairly small. If k is only just greater than unity, as is the case for a reactor using natural uranium as fuel, the system cannot become critical unless its dimensions are large, when P is only slightly less than unity.

1.99. If the nature of the system is such that $k < 1$, then it can never go critical, and a self-sustaining chain reaction is impossible. This is the case, for example, for a *uniform* mixture of natural uranium metal, as fuel, with graphite, as moderator. However, by arranging the fuel and moderator in the form of a lattice, with lumps or rods of natural uranium metal suitably distributed within a matrix of graphite, the value of k becomes slightly greater than unity. By constructing a large reactor, so as to minimize the relative leakage of neutrons, the nonleakage probability is sufficiently close to unity for k_{eff} to be equal to one. Thus, natural uranium-graphite reactors are all large, e.g., the first Chicago pile (CP-1) and its reconstruction (CP-2), the X-10 reactor at Oak Ridge, the Brookhaven reactor, and the Hanford reactors.

1.100. Apart from the value of k, which is, of course, fundamental and which will be considered more fully below, certain other factors determine the critical size. One of these is the shape (or geometry) of the reactor. Since a sphere has the smallest surface for a given volume, it is evident that, for a specified fuel-moderator system, the nonleakage probability will be greatest and, hence, the critical volume (or mass) will be least when the reactor is spherical. It is for this reason that the first successful chain-reacting system (CP-1) was designed to be spherical in shape; however, it became critical before the sphere was completed, and so its actual shape was somewhat like an oblate spheroid.

1.101. The critical size for a specified shape and composition can be decreased by surrounding the chain-reacting region of the reactor, called the *core*, by a blanket of material which scatters the neutrons back into the reactor. Such a blanket is referred to as a *reflector*. By decreasing the neutron leakage, for a core of given size, the reflector increases the nonleakage probability. Consequently, for a given value of the infinite multiplication factor, a reactor with reflector will become critical when the dimensions of the core are appreciably less than those required for a bare reactor, i.e., one without a reflector. The nature of the reflector material is determined largely by the energies of the neutrons responsible for most of the fissions. If these are slow neutrons, then the best reflector will consist of a material containing elements of low atomic weight which do not absorb slow neutrons to any appreciable extent, e.g., heavy water, beryllium (or its oxide), and graphite (carbon).

Energy of Neutrons Causing Fission

1.102. The value of the infinite multiplication factor will depend on the nature of both the fuel and moderator and on their relative proportions. In the first place, the ratio of fissionable material to moderator will determine the neutron energy range within which the majority of the fissions occur. Thus, if there is little or no moderator — including coolant which can act as a moderator — most of the fissions will be caused by the absorption of fast neutrons by the fissionable nuclei. A system of this type is called a *fast reactor;* examples are the Los Alamos Fast Reactor (dismantled in 1953) and the Experimental Breeder Reactor (EBR) at the National Reactor Testing Station, Arco, Idaho. The atomic bomb is a fast reactor in which the chain reaction is so highly divergent that it goes completely out of control.

1.103. When the proportion of moderator is large, slow (thermal) neutrons will be responsible for most of the fissions. For reasons which will become apparent in due course, such systems, referred to as *thermal reactors*, are most common at the present time. Nearly all the reactors now in operation, with the exception of those mentioned in the preceding paragraph, are thermal reactors. An *intermediate reactor*, the Submarine Intermediate Reactor (SIR), wherein fission will result mainly from the absorption of neutrons in the inter-

mediate energy range, between thermal and fast, is being constructed at the Knolls Atomic Power Laboratory, West Milton, N. Y.

Nonfission Capture in Fuel

1.104. The energy distribution of the neutrons in a reactor will influence the infinite multiplication factor k because the proportion of nonfission captures, which affects the value of k, depends on the neutron energy. If there were no parasitic capture of any kind, and every neutron absorbed by the fissionable nuclei resulted in fission, the infinite multiplication factor would be equal to ν, the average number of neutrons liberated *in each act of fission* (§ 1.72). However, not all the neutrons absorbed in the fuel material cause fission, and so the quantity which determines k is η, the average number of neutrons liberated *for each neutron absorbed in the fuel;* this is related to ν by

$$\eta = \nu \times \frac{\text{Neutrons causing fission}}{\text{Total neutrons absorbed in fuel}}. \tag{1.104.1}$$

Since parasitic capture by moderator, coolant, poisons, etc., has been ignored, this gives the maximum value of k for a specified fuel.

1.105. For pure fissionable material, such as pure uranium-233, uranium-235, or plutonium-239, it is convenient to define a quantity α as follows:

$$\alpha = \frac{\text{Neutrons captured in nonfission reactions}}{\text{Neutrons causing fission}} = \frac{\sigma_c}{\sigma_f}, \tag{1.105.1}$$

where σ_c and σ_f are the nuclear cross sections for nonfission capture and for the fission reaction, respectively. Consequently, it follows from equations (1.104.1) and (1.105.1) that

$$\eta = \frac{\nu}{1 + \alpha}. \tag{1.105.2}$$

1.106. The values of α for thermal neutrons are 0.19 for uranium-235 and 0.54 for plutonium-239. Hence, $1/(1 + \alpha)$, which represents the fraction of neutrons absorbed that actually cause fission, is 0.84 for the former and 0.65 for the latter. Using the data for ν in Table 1.3, it is found from equation (1.105.2) that η is about 2.1 for uranium-235 and 1.95 for plutonium-239, for thermal neutrons. Since the cross sections depend on neutron energy, α may be expected to vary with energy; consequently so also will η, assuming ν is essentially independent of neutron energy. For fast neutrons η exceeds 2 for both uranium-235 and plutonium-239. Because η is always appreciably greater than unity (see Table 1.7), it appears that a chain reaction is possible, in principle, for neutrons of all energies, provided the fuel consists entirely of fissionable material.

1.107. If the fuel contains nonfissionable nuclei, in addition, the situation may be quite different. Equation (1.105.2) does not now apply, but equation (1.104.1) is quite general and can be used in such cases, e.g., when the fuel is

natural uranium, consisting of about 0.7 per cent of fissionable uranium-235 and 99.3 per cent of essentially nonfissionable uranium-238.* If N is the number of uranium nuclei (per unit volume) of the isotope indicated by the superscript, then, for any mixture of uranium-235 and -238,

$$\eta_r = \nu \times \frac{N^{235}\sigma_f^{235}}{N^{235}\sigma_f^{235} + N^{235}\sigma_c^{235} + N^{238}\sigma_c^{238}}$$

$$= \nu \times \frac{\sigma_f^{235}}{\sigma_f^{235} + \sigma_c^{235} + \sigma_c^{238}/r} \tag{1.107.1}$$

where r, equal to N^{235}/N^{238}, is the ratio of the two isotopes in the fuel; as before, σ_f and σ_c represent the fission and capture (nonfission) cross sections, respectively. For natural uranium, r is 0.00717 (see Table 1.1) and, using the known cross sections (Table 2.10), η_r is found to be 1.3 for thermal-neutron absorption, but for neutrons of high or intermediate energy, η_r is less than unity (Table 1.7).

TABLE 1.7. AVERAGE NUMBER OF NEUTRONS LIBERATED PER NEUTRON ABSORBED IN FUEL

Neutrons Absorbed	*Uranium-235*	*Plutonium-239*	*Natural Uranium*
Thermal	2.1_1	1.9_5	1.3_2
Fast	> 2	> 2	< 1

As η is the maximum possible value of the infinite multiplication factor in the given circumstances, it is evident that only when the majority of neutrons have thermal energies is a self-sustaining chain reaction possible with natural uranium as fuel.

1.108. By using "enriched" fuel, that is, material in which the proportion of fissionable to nonfissionable nuclei is increased artificially, i.e., by increasing the value of r in equation (1.107.1), η_r can be made to exceed unity even for neutrons in the intermediate energy range. For a fast reactor the fuel must consist largely of fissionable material because collisions with nonfissionable nuclei would cause a decrease in the average neutron energy.

1.109. As stated above, η (or η_r) gives the maximum possible value of the infinite multiplication factor, since it ignores parasitic capture of neutrons. Further, it does not take into account resonance capture by nonfissionable nuclei present in the fuel (cf. § 1.91). These matters will be considered later, since they are not essential to the general discussion given here.

The Critical Equation

1.110. Some interesting qualitative conclusions concerning the critical size of a reactor may be obtained from the following considerations. Since k is the number of neutrons produced in each generation per neutron absorbed, and only one of these neutrons is required to maintain the fission chain when the reactor is critical, it is evident that $k - 1$ is a measure of the relative neutron

* The uranium-238 is fissionable by fast neutrons, and allowance can be made for this (§ 3.131). As stated earlier, uranium-234 is ignored.

leakage in a finite critical reactor. The extent of this leakage will depend on two factors; the first is the dimensions of the reactor, and the second is the average (vector) distance a neutron travels between its birth, as a fission neutron, and its ultimate absorption by a nucleus. This distance is related to a quantity called the *migration length* (§ 3.99).

1.111. It can be seen from general principles that the chance of a neutron escaping will increase with the migration length but will decrease with increasing size of the reactor. Hence, if R represents a linear dimension of the reactor, e.g., radius of a sphere, side of a cube, etc., and M is the migration length, the relative leakage will be determined by the ratio M/R. A more exact treatment, given in Chapter III, shows that it is the square of this ratio that determines the relative neutron leakage, and so it is possible to write for a critical reactor the dimensionally correct (approximate) relationship

$$k - 1 \approx \frac{bM^2}{R^2}, \tag{1.111.1}$$

where b is a numerical quantity which includes a factor for the geometry (or shape) of the reactor. This result is a form of what is called the *critical equation* for a bare reactor. By transposing to

$$R \approx M\left(\frac{b}{k-1}\right)^{\frac{1}{2}}, \tag{1.111.2}$$

the critical equation is seen to relate the size, i.e., the R value, of the critical reactor to the infinite multiplication factor and the migration length, both of which are specific properties of the system.

Reactor Size and Characteristics

1.112. The migration length is generally greatest for slow neutrons because it involves the net distance covered by the neutrons while slowing down, i.e., the slowing down length, as well as the distance traveled as a thermal neutron before capture, i.e., the thermal diffusion length. For fast neutrons, on the other hand, the migration length is roughly equal to the fast diffusion length, since there is virtually no slowing down. It is to be expected, therefore, that the migration length will be smallest for fast neutrons. Hence, it follows from equation (1.111.2) that, for a given value of k, the critical size of a fast reactor is usually less than that of a slow (thermal) reactor. For neutrons of a specified energy — thermal neutrons in particular — the reactor size can be decreased by an increase in k; this may be achieved by using a fuel having a higher value of η (Table 1.7), e.g., by increasing the proportion of fissionable species.

1.113. The foregoing general conclusions may be illustrated, as regards order of magnitude, by considering some examples. For a fast reactor, in which the fuel must consist of essentially pure fissionable material, k may be about 2 and M, which can be roughly estimated on purely theoretical grounds (see example

in § 2.147), is something like 5 cm, i.e., 2 in. The geometrical factor b is equal to π^2 for a sphere, and so R, the critical radius of a bare fast reactor, is found from equation (1.111.2) to be around 15 cm, or 6 in.

1.114. The critical size of a thermal reactor, which depends on the nature of both fuel and moderator, can vary over a considerable range. At one extreme may be considered a reactor in which natural uranium is the fuel and carbon (graphite) the moderator. Here k is not more than about 1.05 and M is at least 50 cm, so that the critical radius is roughly 700 cm, or 23 ft. At the other extreme would be a system consisting of pure (or highly enriched) fissionable material as fuel with ordinary water as the moderator. The migration length of thermal neutrons in water is exceptionally low — about 7 cm — and since k may be about 1.5, at least, in view of the larger value of η, the critical radius of the bare reactor would be roughly 30 cm, or 12 in.

1.115. It might appear from the foregoing that the critical size of a natural-uranium reactor could be decreased by using ordinary water as moderator in place of graphite. Unfortunately, this cannot be done, because capture of slow neutrons by water is so considerable that the infinite multiplication factor is less than unity. Consequently a self-sustaining fission chain reaction is impossible no matter how large the reactor. By enriching the uranium fuel in uranium-235, k can be made greater than unity, because of the accompanying increase in η, and then a critical system with water as moderator can be achieved. Such is the case in the Materials Testing Reactor at Arco, in the Submarine Thermal Reactor (§ 1.22), and in the various "Water Boilers" at Los Alamos Scientific Laboratory, at North Carolina State College, and elsewhere. The cores of all these reactors are relatively small because of the high k and the low value of the migration length of thermal neutrons in water.

1.116. If natural uranium is to be used as the fuel, then an appreciable decrease in size can be achieved by employing heavy water as the moderator. Although the migration length of thermal neutrons in this medium is larger than in graphite, the increase is more than offset by the accompanying gain in the multiplication factor k, because of the much smaller loss of neutrons due to parasitic capture by the moderator. As a result, a critical reactor of natural uranium and heavy water can be smaller than one using graphite as moderator.

TABLE 1.8. TYPICAL REACTOR SYSTEMS

Fuel	Moderator	Neutrons	Coolant	Dimensions (ft)
Natural uranium	Graphite	Slow	Air	25
Natural uranium	Heavy water	Slow	Heavy water	10
Slightly enriched U	Graphite	Slow	Sodium	8
Slightly enriched U	Water	Slow	Water	5
Highly enriched U	Water	Slow	Water	1.5
Highly enriched U	None	Fast	Sodium	1

1.117. The approximate sizes of a number of reactor systems of different types are quoted in Table 1.8.* The dimensions given are for the diameter and the height, assumed to be the same, of a cylindrical reactor. These results, as well as those in the illustrations given above, are intended to indicate an order of magnitude only, since reactor dimensions are affected by the degree of enrichment of the fuel, by the ratio of fuel to moderator, and by the presence of various materials which capture neutrons. It should be recalled, too, that a suitable reflector (see § 1.101) will result in appreciable decrease in the critical size.

Power Output

1.118. In connection with the discussion of reactor size, it may be repeated that, unlike other means of power production, the steady power output of a nuclear reactor is not directly related to its size. The determining factor is the rate of heat removal from the system. If the temperature of the reactor is not to rise continuously, the heat produced by fission must be removed at the same rate as it is being generated. The maximum power output of the reactor then depends on the rate at which heat can be removed. Because of engineering problems, it is more difficult to remove heat from a small reactor than from a large one. Thus, there are, at present, technological limitations upon the design of a stable fast reactor with a high power output.

1.119. For thermal reactors there is another factor which sets a limit upon the power output; this is the accumulation of fission product poisons with large cross sections for the capture of thermal neutrons. The most important of these is xenon-135, and xenon poisoning is of considerable significance in decreasing the multiplication factor. While the reactor is in operation the xenon concentration increases toward an equilibrium value; the rate of its formation, by decay of a fission product, is then just equal to the rate of removal by its own radioactive decay and by neutron capture. When the reactor is shut down, however, the xenon concentration increases steadily for several hours — because it continues to be formed as a decay product — before reaching a maximum, after which it decreases slowly.

1.120. Quite apart from the effect on the multiplication factor of the equilibrium amount of xenon while the reactor is operating, and for which allowance can be made, there is the much more serious problem of the accumulation after shut-down. The growth of the xenon concentration is not important for reactors of low power. But as the power is increased the poisoning may become so considerable that, if the reactor is shut down for an hour or two, it may be necessary to wait a day or more for the xenon to decay to a sufficient extent to permit the reactor to be started again.

1.121. It is seen, therefore, that, whereas the power output of a fast reactor is limited by the rate of heat removal, that of a thermal reactor may be influenced by the xenon poisoning. Consequently, partly as a compromise and partly for

* J. A. Lane and S. McLain, *Chem. Eng. Progress*, **49**, 287 (1953).

other reasons, there has been some interest in intermediate reactors, e.g., in the Submarine Intermediate Reactor (§ 1.22), for power production.

Reactor Control

1.122. Although a sufficient condition for the steady operation of a nuclear reactor is that the effective multiplication factor should be unity, an appreciable power output is possible only if k_{eff} can be made to exceed unity, at least when the reactor is being started, i.e., during start-up. It will be shown in Chapter II that the power output of a given reactor is directly proportional to the neutron density, i.e., number of neutrons per unit volume. The only practical way for this density to be increased up to a desired level is to make use of a divergent fission-chain reaction, for which $k_{eff} > 1$ (§ 1.94).

1.123. When a reactor is constructed, it is always made somewhat larger than the critical size, i.e., it contains more than the critical amount of fuel, so that k_{eff} can be greater than unity. However, until the reactor is ready for operation, the value of k_{eff} is maintained less than unity, often by the introduction of a strong neutron absorber, called a *control rod*. Every reactor, even when shut down, has a small neutron density, which may be due to spontaneous fission (§ 1.69), to fission caused by neutrons in cosmic rays, or to other sources of neutrons present in the reactor. As long as the system is subcritical, i.e., $k_{eff} < 1$, the neutron density will be quite small and will remain constant. By withdrawing the control rods, so that $k_{eff} > 1$, the divergent chain reaction causes the neutron density to increase, more or less rapidly, until the desired level, corresponding to the required power output, is attained. The control rods are then inserted so as to make k_{eff} equal to unity; the chain now becomes stationary and the power output remains constant.*

1.124. A change in power output can be readily caused either by making $k_{eff} > 1$ to increase the power or $k_{eff} < 1$ to decrease it, for a short time, and then restoring the conditions to $k_{eff} = 1$ again. To shut down the reactor, the absorbing rods are inserted to make k_{eff} considerably less than unity; the convergent chain reaction then causes the neutron density and the power output to decrease to a negligible level.

1.125. The procedure just described, whereby a neutron absorber is used to control the reactor, is based on the ability of the absorber to increase or decrease the k factor of the system, the nonleakage probability P being unaffected. This method is suitable for thermal and intermediate reactors, since good or moderately good absorbers are available for neutrons in these ranges. For fast reactors the use of absorbers is, however, not practical, since absorption cross sections are then very small. In these reactors the control principle used is to keep k more or less constant while P is varied. This is achieved by moving parts of the

* Increase of temperature and accumulation of poisons cause variations which need not be considered here.

reactor core or reflector, thus increasing or decreasing the neutron leakage as required for proper control.

Time Behavior of Reactor

1.126. The rate at which the neutron density (or power) of a given reactor will increase or decrease depends on the amount by which k_{eff} is greater or less, respectively, than unity. An approximate treatment of this subject will be given here, but it will be developed more rigorously in Chapter IV. Suppose the neutron density in a reactor is such that n neutrons are present per unit volume at the beginning of a given generation. All these neutrons will either be absorbed or will leak out, and at the end of the generation nk_{eff} neutrons will be produced, in accordance with the definition of the effective multiplication factor (§ 1.94). Hence the number of neutrons per unit volume gained in the generation is $n(k_{eff} - 1)$. If l is the generation time (or neutron lifetime), i.e., average time between successive neutron generations in the system under consideration, it follows that

$$\frac{dn}{dt} = \frac{n(k_{eff} - 1)}{l} = \frac{nk_{ex}}{l}, \tag{1.126.1}$$

where k_{ex}, called the *excess multiplication factor*, is defined by

$$k_{ex} \equiv k_{eff} - 1, \tag{1.126.2}$$

so that it is a measure of the departure of the reactor from criticality. Upon integration of equation (1.126.1), the result is

$$n = n_0 e^{t(k_{ex}/l)}, \tag{1.126.3}$$

where n_0 is the initial neutron density and n is that after the lapse of time t. The ratio l/k_{ex} is called the *reactor period* and is represented by T, i.e.,

$$T = \frac{l}{k_{ex}}, \tag{1.126.4}$$

so that

$$n = n_0 e^{t/T}. \tag{1.126.5}$$

The reactor period is thus seen to be the time required for the neutron density to change by a factor of e; hence it is sometimes called the e-folding time. The larger the period, the more slowly does the neutron density change with time. Under normal operating conditions, l is constant for a given reactor, and so the period is dependent upon the value of k_{ex}, i.e., upon the departure from criticality.

1.127. Equation (1.126.5) gives the time rate of change of neutron density for either positive or negative values of k_{ex}, that is, for $k_{eff} > 1$ or < 1, respectively. Thus, the neutron density will either increase or decrease exponentially, depending on whether k_{eff} is greater or less than unity. The rate of change is seen to be inversely related to the reactor period, so that for a given value of l, the generation time, the neutron density will increase (or decrease) more rapidly the greater the amount whereby k_{eff} exceeds (or is less than) unity. It should be noted from equations (1.126.3) and (1.126.4) that, as long as $k_{eff} \neq 1$ so that k_{ex} is not zero, the neutron density (or power output) of a chain reactor cannot remain constant; it must either increase if $k_{eff} > 1$ or decrease if $k_{eff} < 1$. Only when $k_{eff} = 1$ does the neutron density and power output remain steady. This is in agreement with the general conclusions reached earlier.

Effect of Delayed Neutrons

1.128. In increasing or decreasing the neutron density, for the purpose of reactor control, the generation time is a highly significant quantity. The average time elapsing between the production of a fission neutron and its ultimate capture as a thermal neutron, for example, is about 10^{-3} sec in a natural-uranium reactor. Since the prompt fission neutrons are released almost instantaneously, the generation time, as far as these neutrons are concerned, is 10^{-3} sec. However, as already seen (§ 1.74), an appreciable fraction of the fission neutrons are delayed, and so the *effective* generation time (or neutron lifetime) is greatly increased.

1.129. The mean lives (emission times) of the five groups of delayed neutrons range from 0.6 sec to 80 sec (see Table 2.11). By weighting the values appropriately, according to the fractions in each group, the mean delay time, averaged over all the delayed neutrons, is about 0.1 sec for the fission of uranium-235 (§ 4.22). The average effective generation time is, thus, about 0.001 + 0.1 sec; the first term is the average time elapsing between release of a neutron of one generation and its capture in a fission process, whereas the second is the average time elapsing between fission and the effective complete release of the neutron of the next generation.

1.130. The value of l to be used in equation (1.126.3), when uranium-235 undergoes fission, is therefore about 0.1 sec, so that

$$n \approx n_0 e^{10tk_{ex}}, \tag{1.130.1}$$

with t in sec, provided the delayed neutrons are rate-determining. If k_{eff} is 1.005, i.e., $k_{ex} = 0.005$, then the neutron density will increase by a factor of $e^{0.05}$, i.e., about 1.05, per sec. By the end of 100 sec, the increase will be e^5, i.e., about 150-fold. This is a relatively slow and controllable rate of increase, since in the start-up of a reactor it is often necessary to increase the neutron density by a factor of 10^{10} or more. It should be noted that, if it were not for the delayed neutrons, l would be 10^{-3} sec for a natural-uranium reactor; the neutron density, for $k_{eff} = 1.005$, would then increase by a factor of e^5, i.e., 150-fold, per sec. Such a rapid rate of increase in the power output would make the reactor go out of control in a few seconds.

1.131. Although the mean delay time of the neutrons determines the rate of neutron growth in the example considered above, it must be borne in mind that

this is not always the case. Suppose that, in general, β is the fraction of the fission neutrons which are delayed, so that $1 - \beta$ is the fraction of prompt neutrons. The effective multiplication factor may then be considered as consisting of two parts: one, equal to $k_{\text{eff}}(1 - \beta)$, representing the prompt neutron multiplication factor, and the other, equal to $k_{\text{eff}}\beta$, being due to delayed neutrons. If, in the operation of a reactor, the quantity $k_{\text{eff}}(1 - \beta)$ is just less than (or equal to) unity, then the rate of increase in the number of neutrons from one generation to the next will be determined essentially by the delayed neutrons. Since β is about 0.0075 for the thermal fission of uranium-235,* this condition can be realized, provided the effective multiplication factor is kept between 1 and $1/(1 - \beta)$, i.e., between 1 and 1.0075. When such is the case, equation (1.130.1) will be applicable; the neutron density and power level of the reactor can be increased relatively slowly and adequate control is possible.

1.132. When the value of $k_{\text{eff}}(1 - \beta)$ is exactly unity, i.e., $k_{\text{eff}} = 1.0075$ for uranium-235 fission, the reactor is described as *prompt critical*, since it will be critical on prompt neutrons alone. If $k_{\text{eff}}(1 - \beta)$ is greater than unity, i.e., $k_{\text{eff}} > 1.0075$, the fission chain will be divergent due to the release of prompt neutrons, irrespective of those delayed. In these circumstances, the l to be used in equation (1.126.3) is the generation time (or lifetime) of the prompt neutrons, i.e., about 10^{-3} sec for a natural-uranium reactor. The combination of $k_{\text{ex}} > 0.0075$ and the small value of l means that the neutron density will increase at a very rapid rate. In this condition, a reactor is difficult to control and so it must be avoided in practice.

1.133. Just as the delayed fission neutrons affect the rate of increase of neutron density when the effective multiplication factor exceeds unity, so they influence the rate of decrease when the reactor is made subcritical, i.e., when it is being shut down. The delayed neutrons continue to be emitted for some time, and so maintain a fission rate that is considerably higher than would be the case if all the fission neutrons were prompt. The ultimate rate at which the neutron density in a reactor decreases after shut-down is essentially independent of the value of k_{eff} and is determined by the most delayed group of fission neutrons, i.e., by those with a mean life of 80 sec. The rate of decrease is then given by equation (1.126.5) with the period T equal to -80 sec.†

1.134. The numerical values in the foregoing discussion have referred in particular to thermal fission in natural-uranium systems. For enriched thermal reactors, such as the Materials Testing Reactor and the Submarine Thermal Reactor, the mean delay time of the fission neutrons is the same as given above, so that, provided the reactor is below prompt critical, the time behavior will be much the same as for a natural-uranium reactor. However, the prompt-neutron

* For plutonium-239 the fraction β of delayed fission neutrons is about 0.0036.

† In reactors using heavy water or beryllium as moderator or reflector, the rate of shut-down is even slower, due to the liberation of neutrons by the action of gamma rays from the fission products.

generation time is of the order of 10^{-4} sec in these enriched reactors, so that if the multiplication factor exceeds the prompt-critical value, the neutron density and power level will increase more rapidly than in the natural-uranium system.

1.135. In a fast reactor, the prompt generation time (or neutron lifetime) is much less than in a thermal or intermediate reactor; it may be 10^{-7} sec or less. Consequently, when $k_{\text{eff}}(1 - \beta) > 1$, the neutron density will increase with extreme rapidity. However, provided $k_{\text{eff}}(1 - \beta)$ is less than unity, so that the system is below prompt critical, a fast reactor, e.g., the Experimental Breeder Reactor, is just as controllable as a thermal reactor, because of the delayed neutrons.

REACTORS FOR PRODUCTION AND POWER

Production Reactors

1.136. Apart from the fission products, which have certain uses, a nuclear reactor produces two important commodities, namely, energy and neutrons. The discussion so far has been based on the supposition that the purpose of a reactor is the release of nuclear energy. While this must represent the ultimate objective of most reactors,* there are some reactors, such as those at Hanford, in which the energy produced by fission is largely wasted. In these reactors the neutrons accompanying the fission process are used to convert an essentially nonfissionable material (uranium-238) into one (plutonium-239) that is fissionable by neutrons of all energies. Such reactors have been called *production reactors* or *converters;* their prime purpose is to make substances that can be used for the liberation of nuclear energy, either in a controlled manner for power production or in an uncontrolled manner in atomic bombs.

1.137. The absorption of one neutron in fissionable material results in the liberation of at least two neutrons (Table 1.3). One of these is required to maintain the chain reaction, and so there is at least one additional neutron available for other purposes. By having a fairly large reactor, so as to reduce the loss of neutrons by leakage, and keeping parasitic capture to a minimum, by eliminating, as far as possible, all extraneous substances with large capture cross sections, an appreciable proportion of the available neutrons can be captured in uranium-238 or thorium-232. As seen earlier (§§ 1.59, 1.61), the products of these radiative capture reaction undergo two stages of radioactive decay and are thereby converted into the fissionable plutonium-239 or uranium-233, respectively. The uranium-238 and thorium-232, which by themselves cannot sustain a chain reaction, but which can be transformed into nuclei capable of doing so, are called *fertile materials.*

1.138. In a reactor using natural uranium as fuel, the capture of resonance (intermediate energy) and thermal neutrons by uranium-238 is unavoidable.

* The exceptions are reactors used for experimental and test purposes, and for the production of certain isotopes.

It is seen from the neutron balance presented in § 1.92 that, for every 40 neutrons causing fission of uranium-235, about 43 neutrons are captured by uranium-238, moderator, etc.* If the parasitic capture is small, then it is possible for the same number of neutrons to be absorbed in fission of uranium-235 as are captured by the fertile uranium-238 nuclei. Each of the latter will, after neutron capture and radioactive decay, be converted into a plutonium-239 nucleus, and so, for every nucleus of uranium-235 undergoing fission, there will, in due course, be produced a fissionable plutonium-239 nucleus.

1.139. The conversion efficiency, i.e., the ratio of fissionable nuclei produced to those lost, in the hypothetical case considered above is just 100 per cent — one new fissionable nucleus formed for each one destroyed. However, if there is a considerable amount of leakage and parasitic capture, the conversion efficiency would be less than 100 per cent. On the other hand, it could conceivably be more, but not much more, than 100 per cent, since 2.1 neutrons are produced, on the average, for each thermal neutron absorbed in uranium-235 (Table 1.7), leaving 1.1 neutrons available for nonfission capture. Assuming the conversion efficiency to be 100 per cent, it follows, from the statement in § 1.87, that a production reactor will produce about 1 gram of plutonium per day when operating at a power of 1 megawatt.

Separation of Fissionable Material

1.140. It is legitimate at this point to ask: What is the purpose of a reactor which does not utilize the energy released in fission, but merely uses one fissionable material to produce a roughly equal amount of another? The answer to this question is bound up with the historical development of the atomic energy project, as well as with other, more fundamental, matters connected with the properties of the various fissionable nuclei.

1.141. For use in atomic weapons it was necessary to obtain fissionable material in a relatively pure state. For this purpose the uranium-235 in natural uranium is of no value unless it can be separated from the heavier isotope, uranium-238. Because the isotopes of an element have essentially identical chemical properties, physical methods of separation must be used. Various procedures have been tried for separating the isotopes of uranium. While simple in principle, they all involve very large plants and a great expenditure of power to obtain appreciable quantities of uranium-235. In other words, the separation of uranium-235 from natural uranium is slow and costly.

1.142. The process which has proved most practical is the gaseous-diffusion method, as originally employed at Oak Ridge, Tennessee. It depends upon the fact that the rate of diffusion of a gaseous substance is inversely proportional to the square root of its molecular weight. Consequently, in a mixture of two

* In the same system 7 neutrons are captured in a nonfission (radiative capture) reaction with uranium-235. No particular use has yet been found for the product, uranium-236.

isotopic species the lighter one will diffuse more rapidly than the heavier, and a partial separation is possible. For the separation of uranium isotopes the gas used is uranium hexafluoride (UF_6), so that the molecular weight of the lighter isotopic form, with uranium-235, is 349, whereas that of the heavier form is 352. The ratio of the diffusion rates is therefore $\sqrt{352/349}$, i.e., 1.0043. Since this number is so close to unity, the degree of separation achieved in a single diffusion stage is very small, and several thousand stages are required to obtain uranium-235 in a relatively pure form.

1.143. In order to avoid complete dependence upon physical methods of separating fissionable material for use in atomic weapons, it was decided in 1942 to proceed with the alternative method of using the unseparated uranium-235, in natural uranium, to produce fissionable plutonium-239 in a reactor, as already described. The apparent advantage of this procedure is that, since plutonium and uranium are different elements, they have different chemical properties and so they should be separable by more or less familiar chemical methods. However, for several reasons, this separation is by no means simple. Because plutonium-239 is fissionable by slow neutrons, it will compete with the uranium-235 as it accumulates in the reactor. There is obviously no purpose in destroying plutonium nuclei in order to produce others, and so the process must be stopped, and the plutonium separated, when only a relatively small proportion is present in the system.

1.144. Apart from the difficulty of efficiently separating small quantities of plutonium from large amounts of uranium, it must be remembered that the mixture will contain fission products consisting of more than 30 different elements. The plutonium must be separated from these as well as from the uranium. The chemical problems involved are difficult enough, but they are greatly aggravated by the fact that the fission product mixture is highly radioactive and represents a serious hazard to personnel. The complex separation processes must therefore be operated, and the plants maintained, by remote control.

1.145. It is apparent that neither separation of uranium-235 nor of plutonium-239 is a simple matter. Nevertheless, both methods for the production of fairly pure fissionable material are being used, and even extended, at the present time.

1.146. For the production of useful power a reactor should be moderately compact and should have some flexibility in operation. These requirements can be met by the use of enriched fuel, i.e., one containing a larger than normal proportion of fissionable nuclei. Uranium-235 is probably to be preferred to plutonium-239 for the enrichment of reactor fuel. In the first place, fabrication of fuel elements is less of a health hazard than for plutonium and, in the second place, the larger fraction of delayed fission neutrons, about 0.0075 as compared with 0.0036 from plutonium-239, permits somewhat wider scope in the control

of a reactor in which uranium-235 is the fissionable substance. Plutonium is, nevertheless, destined to play an important role in the development of nuclear energy, as will be seen in the next section.

Conversion and Breeding

1.147. Because uranium-235 is the only fissionable species occurring in nature to any extent, this must serve, initially, to maintain the chain reaction in production reactors. Excess fission neutrons are then captured by fertile uranium-238, which is thereby converted into fissionable plutonium-239. Since natural uranium contains only 0.7 per cent of uranium-235, it is evident that, in due course, almost all the available uranium-235 will be consumed and will be replaced by an approximately equivalent amount of plutonium-239. At this time, however, nearly 140 times as much of nonfissionable uranium-238 will remain. If this is to be utilized for power production, then reactors of the future must use plutonium-239 to maintain the fission chain and be capable of regenerating the same species at the expense of uranium-238. In the long run, therefore, it is reactors of this type which will be of basic significance.

1.148. If such a reactor were to regenerate the same amount of plutonium-239 as it loses by fission, i.e., if the conversion efficiency were 100 per cent, the total quantity of fissionable material available for energy production would remain constant. Provided there were no losses, it would then be possible, in principle, to make use of virtually all the available fertile material. If, however, the conversion efficiency were less than 100 per cent, the stockpile (or inventory) of fissionable material would gradually diminish and ultimately it would be exhausted. Nuclear power production through fission would then cease to be feasible, even though large amounts of fertile uranium-238 remained.

1.149. The prospect of greatest interest is that of a regenerative reactor with a conversion efficiency exceeding 100 per cent, i.e., more fissionable material would be produced than is consumed in maintaining the fission chain. The process whereby a fissionable species is utilized as a source of neutrons to produce more nuclei of its own kind than are used up is called *breeding*. If breeding is possible, then the stockpile of fissionable material can be steadily increased until essentially no more uranium-238 is left.

1.150. The foregoing discussion has referred, in particular, to uranium-238 as the fertile material, but the arguments apply equally to thorium-232. While uranium-235 is available, thorium can be converted into fissionable uranium-233. If the thorium-232 supplies in nature are to be utilized as a source of power, then eventually reactors must be developed in which uranium-233 serves to maintain a fission chain and at the same time supplies neutrons for its regeneration from thorium-232. To make full use of the latter, the conversion efficiency must be equal to or greater than 100 per cent.

Utilization of Fertile Material

1.151. It was seen in § 1.104 that, for every neutron absorbed by the fissionable material, there are produced, on the average, η fission neutrons. Of these, one neutron is required to maintain the chain reaction. Suppose that L is the number of neutrons, on the average, lost by leakage and by parasitic capture of all kinds (other than by the fissionable material); then $\eta - 1 - L$ neutrons will be available for capture by fertile nuclei to produce fissionable material. This is consequently the number of new fissionable nuclei produced for each such nucleus consumed (in both fission and nonfission reactions), and so it is called the conversion ratio C; thus,

$$C = \eta - 1 - L. \tag{1.151.1}$$

It should be noted that, in this expression for the conversion ratio, η refers to the fissionable species, e.g., uranium-235, and not to that of the fuel material as a whole, e.g., natural uranium.

1.152. Suppose N nuclei of fissionable material, e.g., plutonium-239, are consumed in a conversion reactor; then NC nuclei of the same species will be regenerated at the expense of the same number of fertile, i.e., uranium-238, nuclei. If the resulting NC nuclei of plutonium-239 then undergo fission in a reactor of the same type, NC^2 nuclei will be obtained, and so on in succeeding stages. The total amount of fissionable material produced and, hence, of fertile material consumed will then be equal to the sum of NC, NC^2, NC^3, etc.; it follows, therefore, that

$$\text{Fertile nuclei converted to fissionable nuclei} = NC + NC^2 + NC^3 + \cdots. \tag{1.152.1}$$

If the conversion ratio C is equal to or greater than unity, i.e., if the conversion efficiency is 100 per cent or more, the series in equation (1.152.1) is divergent and there is no theoretical limit to the quantity of fertile material that can be changed into a fissionable product. Thus, as concluded from the qualitative argument given above, it should be possible, in principle, to utilize all the available fertile material for power production.

1.153. Suppose, however, that C is less than unity, i.e., the conversion efficiency is less than 100 per cent; the series in equation (1.152.1) is then convergent and after a large number of stages the sum approaches a finite limiting value. In these circumstances, i.e., $C < 1$,

$$\text{Limiting conversion of fertile nuclei} = \frac{NC}{1 - C}.$$

If, for example, the conversion efficiency is 90 per cent, so that C is 0.9, the limiting amount of fertile material which can be converted into fissionable material is only $9N$. This means that if a reactor is started with 1 pound of plutonium-239 (or of uranium-235), it should be possible eventually to convert

9 pounds of uranium-238 into plutonium, assuming 90 per cent conversion efficiency at each stage.

1.154. The amount of fissionable material available after x stages is NC^x, and if $C < 1$ this quantity decreases steadily as the number of stages increases. As stated earlier, the inventory of fissionable material will then decrease in the course of time if the conversion efficiency is less than 100 per cent. If C is unity, then NC^x is equal to N at all stages, and the stockpile remains constant, as long as fertile material is available for conversion. The condition for breeding is ■ conversion efficiency exceeding 100 per cent, i.e., $C > 1$. In these circumstances, the fissionable inventory increases from one conversion stage to the next.

Doubling Time and Breeding Gain

1.155. For practical purposes, the effectiveness of a breeder reactor in increasing the quantity of fissionable nuclei is determined by the *doubling time;* this is the time, usually expressed in days, required for the fissionable material in the given reactor to double itself. The shorter the doubling time, the more rapidly can the fissionable inventory be increased.

1.156. The doubling time is related to the breeding gain and the specific power of the reactor. The *breeding gain*, G, is defined as the number of fissionable nuclei gained for each such nucleus consumed. Since C is the number of fissionable nuclei formed for each fissionable nucleus lost, it follows that

$$G = C - 1 = \eta - 2 - L, \tag{1.156.1}$$

using equation (1.151.1) for C. The necessary condition for breeding is that $C > 1$, and so the breeding gain, as defined above, must be positive. The number of fissionable nuclei (or grams) gained per day is equal to G multiplied by the number of nuclei (or grams) consumed per day, i.e.,

$$\text{Grams gained per day} = G \times \text{Grams consumed per day.}$$

Thus, by inversion of both sides,

$$\text{Days required to gain 1 gram} = \frac{1}{G \times \text{Grams consumed per day}}.$$

If the reactor contains 1 gram of fissionable material, the time required to gain 1 gram becomes the doubling time, and so

$$\text{Doubling time in days} = \frac{1}{G \times \text{Grams consumed per day per gram}}. \tag{1.156.2}$$

1.157. Let p be the specific power of the reactor in watts per gram, i.e., the power in watts per gram of fissionable material in the reactor.* Then, since the fission of 1 gram of material per day produces 10^6 watts of power (§ 1.87), it

* The specific power is frequently expressed in kilowatts per kilogram of fissionable material, which is, of course, the same as watts per gram.

follows that

$$\text{Grams } \textit{fissioned} \text{ per day per gram} = \frac{p}{10^6}. \tag{1.157.1}$$

By equation (1.105.1), α is the ratio of neutrons captured in nonfission reactions to those causing fission; hence, $1 + \alpha$ is the ratio of fissionable nuclei consumed (in nonfission and fission reactions) to those undergoing fission. It follows, therefore, from equation (1.157.1) that

$$\text{Grams } \textit{consumed} \text{ per day per gram} = \frac{p(1+\alpha)}{10^6}. \tag{1.157.2}$$

Consequently, upon combining equations (1.156.2) and (1.157.2) the result is

$$\text{Doubling time} = \frac{10^6}{Gp(1+\alpha)} \text{ days}, \tag{1.157.3}$$

so that a high breeding gain and a large specific power will favor the growth of the fissionable stockpile.*

Breeding Possibilities

1.158. It was seen above that the basic requirement for breeding is that the conversion ratio C should be greater than unity or, in other words, that the breeding gain G should be positive. According to equation (1.156.1), the breeding gain is equal to $\eta - 2 - L$ and so the condition for breeding is

$$\eta - 2 - L > 0$$

or

$$\eta > 2 + L.$$

If the losses are made as small as possible, so that L is negligible, the absolute minimum requirement for breeding is

$$\eta > 2,$$

that is, more than two neutrons must be available, on the average, for every neutron absorbed in the fissionable material. One of these neutrons is needed to maintain the fission chain and the remainder, more than one, can be captured by fertile nuclei to produce fissionable material with a conversion efficiency exceeding 100 per cent.

1.159. An examination of Table 1.7 shows that a plutonium breeder depending on thermal-neutron fission is not feasible, since η is only 1.95. However, if fissions are caused by fast neutrons, then breeding might be possible. It is primarily for this reason that the so-called Experimental Breeder Reactor was designed as a fast reactor. Strictly speaking, the EBR is not a breeder, since it uses uranium-235 as fuel and uranium-238 as fertile material. The fission-

* It should be noted that the doubling time calculated refers to the time in the reactor. To this should be added the time required for chemical processing and extracting the fissionable material.

able product is, therefore, plutonium-239. Since η for absorption of fast neutrons by uranium-235 is >2, a conversion efficiency of at least 100 per cent is theoretically possible and has in fact been demonstrated. There is thus every reason to believe that a reactor of similar design, using fast neutrons, will function as a true breeder with plutonium-239 as fuel and uranium-238 as the fertile material.

1.160. Although no data for uranium-233 are included in Table 1.7, it has been stated that η is greater than 2 for thermal (and also for fast) neutron absorption. This means that a thermal breeder, having uranium-233 as the fissionable species and thorium-232 as the fertile material, might be possible. Several preliminary designs for a breeder reactor of this general type have been made and at least one is at present in the stage of active development (see § 13.105).

Single- and Dual-purpose Reactors

1.161. A single-purpose reactor may be designed for the sole (or main) purpose of converting fertile into fissionable material. At the present time, such reactors are wasteful, although they may, nevertheless, be justifiable. In the production reactors at Hanford, for example, there is a considerable release of nuclear energy. This amounts to about 10 million kw-hr of heat for every pound of uranium-235 undergoing fission (see Table 1.6) or, roughly, for every pound of plutonium produced, assuming 100 per cent conversion efficiency. Nearly all of this energy is lost; it serves merely to raise the temperature of the Columbia River, the water from which is used for cooling the reactors.* This is one reason why there is considerable interest in dual-purpose reactors which will produce plutonium-239 for weapons and electrical power for industry (see § 1.23). Such reactors would use both the energy and the neutrons released in fission — the energy for power production and the neutrons, in excess of those required to maintain the fission chain, for the conversion of uranium-238 into plutonium-239.

1.162. A power-only reactor, using highly enriched fuel, would be wasteful in neutrons; since only one neutron is needed for fission, to carry on the chain, the remaining fission neutrons either escape or are lost in useless nuclear reactions. The operation of such a reactor would be accompanied by a decrease in the stockpile of fissionable material. Ultimately, when the inventory is large, highly enriched reactors for power production will be more common. In the meantime, power reactors should be designed so as to achieve conversion of fertile into fissionable material with an efficiency approaching 100 per cent, at least. The ideal system would be the power-breeder reactor which would utilize the energy of fission for electrical power and the neutrons to convert fertile into fissionable material with an efficiency exceeding 100 per cent.

* A small part of the fission energy is used to heat some of the buildings at Hanford.

1.163. The purpose of the Experimental Breeder Reactor was to obtain information which would be useful for the design of power-breeders. This reactor has already produced useful amounts of power and has achieved a conversion efficiency of at least 100 per cent, as stated above. It consequently represents a significant step in the development of nuclear energy.

1.164. It should be noted that there is an important difference between a dual-purpose reactor, referred to earlier, which is required to produce plutonium-239 suitable for weapons and a power reactor which merely regenerates (or breeds) fissionable material for further use in reactors. For various reasons, the latter type would be simpler to design and cheaper to construct and operate. Unfortunately the international situation makes it necessary to place emphasis on the former at this time.

Processing Efficiency and Breeding

1.165. It is important to point out that the success or failure of breeding is ultimately bound up with the efficiency of the process used for separating the fuel from the fertile material, e.g., uranium-233 from thorium, or plutonium-239 from uranium, and the fission products. The quantity G defined above may be regarded as the *potential* breeding gain, but the *actual* breeding gain is the net surplus of fissionable material available per nucleus of fuel used up in fission or lost in the separation process. It is very unlikely that the process for separating or extracting the required material from a complex mixture will be 100 per cent effective, and some will inevitably be lost in waste solutions, by adherence to the walls of vessels, as well as in other ways. If the fuel is in the metallic form, as is frequently the case, there are also inevitable losses in the metallurgical process and in fabrication of the fuel units.

1.166. Such losses could have a serious effect on the over-all breeding gain, as the following figures will show. Suppose, in a particular case, that η is 2.5 and L is 0.2; hence, by equation (1.151.1), the potential breeding gain is 0.3. Because of the accumulation of fission products, which capture neutrons, and for other reasons, it will in general be necessary to process the material after the fuel has been only partly consumed. Suppose that 1 per cent has been used up when processing is desirable, and that 0.1 per cent of the fuel is lost in each reprocessing. This means that 0.1 fuel nuclei will be lost in this manner for each nucleus undergoing fission. The net surplus of fissionable material is now 0.2 for 1.1 fuel nuclei used up in fission and in process losses; the actual breeding gain is consequently $0.2/1.1 = 0.18$, compared with the potential value of 0.3, a reduction of 40 per cent.

ASPECTS OF REACTOR DESIGN

General Characteristics of Reactors

1.167. The whole subject of nuclear reactor design is still in its infancy, and nearly all existing reactors differ from one another in certain respects, since they

are mostly intended to serve different purposes. There are, nevertheless, a number of general features which all reactors possess in common, to a greater or lesser extent. In outline, a reactor consists of an active core in which all, or nearly all, of the nuclear fissions occur, and in which most of the energy is released as heat. The core contains the fissionable material, in a suitable form, and also more or less moderator; the relative amounts and nature of the fuel and moderator determine the neutron energy range in which most of the fissions occur. The core is then surrounded by a reflector of a material depending on the energy of the neutrons causing fission. The combination of core and reflector must, of course, be such that it is capable of maintaining a fission chain reaction at the required power level. Control is achieved either by rods, containing a strong neutron absorber, or by moving parts of the core or reflector so as to permit (or prevent) escape of neutrons (Fig. 1.2).

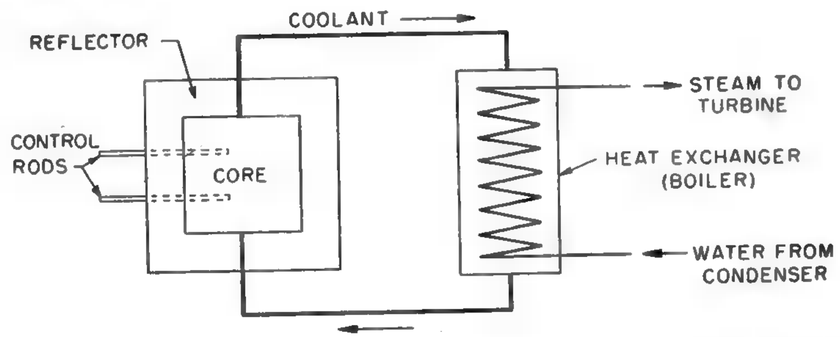


FIG. 1.2. Diagrammatic representation of reactor and heat exchanger

1.168. In a converter (or breeder) type reactor the fertile material may be included in the core, e.g., when natural uranium is the fuel, or it may be in the form of a blanket surrounding the core. In the latter case the blanket may also serve as a reflector, to some extent at least. In a thermal-neutron reactor the particular arrangement chosen is determined by the circumstances, e.g., main purpose of reactor, nature of fuel and fertile material, etc. If fissions are to be caused by fast neutrons, however, as in a plutonium breeder, the fissionable species in the core must not be greatly diluted; in this event the fertile material is mainly incorporated as a blanket.

1.169. The operation of the reactor at appreciable power levels depends on the ability to remove the heat produced by fission as fast as it is liberated. This involves the use of a coolant which must circulate throughout the reactor in such a manner as to maintain a temperature distribution that is as uniform as possible. The design of the reactor core is intimately bound up with the problems of heat transfer, for it is the rate of heat removal which determines the power output. The method used for cooling the reactor is thus of fundamental importance.

1.170. The nuclear reactor is, in essence, a furnace in which heat is produced from the energy released in fission, but until recently it was considered impractical to use a reactor directly as a boiler to vaporize a liquid, such as water, because of the instability due to bubble formation. The design of boiling reactors is now under investigation, but in most power reactor designs the energy is taken up by a coolant and then transferred to water in a suitable heat exchanger. The resulting steam is then used to generate power in some conventional manner, e.g., by means of a turbine. Thus, the heat exchanger associated with a nuclear reactor plays the same role as a boiler in a normal power plant (see Fig. 1.2).

REQUIREMENTS FOR REACTOR MATERIALS

1.171. In general, it would appear that the fundamental principles of reactor design are not very complicated and are well understood. However, it is in the translation of these principles into practice that numerous difficulties are encountered. These are associated largely with the capture of neutrons and with the effect of simultaneous high temperatures, intense gamma radiation, and neutron bombardment on structural and other materials. Another difficulty arises from the intense radioactivity of the fission products which accumulate as the reactor operates and from the radioactivity induced by neutrons in various materials.

1.172. A nuclear reactor presents the possibility of high thermodynamic efficiency in the utilization of heat, because it is liberated at high temperatures. Unfortunately, a limitation is imposed by the nature of both the structural materials and the coolant. These substances must not only be able to withstand the temperature, but they must also have low cross sections for neutron absorption. Any loss of neutrons must be compensated by an increase in the amount of fissionable material if a chain reaction is to be maintained. Further, neutron bombardment causes more or less permanent displacement of the atoms from their normal positions in the crystal lattice. As a result of this disruption of the internal structure, the physical properties of the material may be seriously affected. This phenomenon must be taken into account in the choice of various structural materials as well as in the design of the fuel elements.

1.173. Like the structural materials, the coolant must not absorb neutrons to any appreciable extent and it must be stable at high temperature and to the action of the intense neutron and gamma radiation fields. Very few, if any, organic liquids can meet these specifications. In addition, it should have a high boiling point, if liquid, and be efficient for the transfer of heat. Metals or alloys of low melting point, such as sodium or sodium-potassium alloy (NaK), appear to be promising as coolants. If the coolant captures some neutrons, a radioactive product may be formed; in a power reactor the fluid circulates between the reactor and heat exchanger, and so there is a possibility that the latter may become a hazard to operating personnel.

1.174. In Table 1.9 is given a summary of the possible materials having properties which permit their use as fundamental components of a reactor, namely,

TABLE 1.9. POSSIBLE REACTOR MATERIALS

Nuclear Fuel	Moderator	Reflector	Coolant	Structure
Natural uranium	Heavy water, beryllium, beryllium oxide, or graphite	Any of the moderators	Air, hydrogen, helium, nitrogen, carbon dioxide, heavy water, or ordinary water	Aluminum or zirconium
Enriched uranium	Same as above or ordinary water	Any moderator	Same as above or sodium (or NaK alloy)	Aluminum, zirconium, or stainless steel
Highly enriched U-233, U-235, or Pu-239	None for fast reactor	Uranium-238, thorium-232, or heavy element	Sodium (or NaK) or mercury	Stainless steel

fuel, moderator, reflector, coolant, and structural material. Although many different combinations are possible, certain limitations may be noted. If natural uranium is the fuel, then only a large thermal reactor is possible; the practical coolants are one of the gases or water. The reflector would have to be preferably heavy water, beryllium or its oxide, or graphite. The other substances mentioned as reflectors, namely, uranium-238, thorium-232, or other heavy element, could be used only for a fast reactor. This would need highly enriched uranium-233, uranium-235, or plutonium-239 as fuel, and no moderator. Mercury could be used as coolant, as it was in the Los Alamos Fast Reactor, since the capture cross section for fast neutrons is quite small. The considerable absorption of slow neutrons virtually rules out mercury as a coolant for thermal reactors. Water could be used for the latter, but not for the fast reactor, since it is a good moderator and would slow down the neutrons. The use of the various materials in a number of reactors is summarized in Table 1.10.

Reactor Types

1.175. It is clear that the choice of reactor components is determined, to some extent, by the energy of the neutrons. Therefore this represents one of the first decisions that must be made in connection with the design of a reactor for a particular purpose. A fast reactor has the advantage of relatively small parasitic losses because of the low cross sections for neutron capture by impurities, including fission products. However, the power output is restricted because of the small size and the difficulty of heat removal. Thermal reactors are simpler to control and can be designed to have a high power density. Water, under

TABLE 1.10. SUMMARY OF REACTOR CHARACTERISTICS

Reactor and Location	Type	Moderator	Reflector	Coolant
Natural Uranium Fuel				
CP-1, CP-2 (Argonne National Laboratory)	Thermal, heterogeneous	Graphite	Graphite	Ambient air
X-10 (Oak Ridge National Laboratory)	Thermal, heterogeneous	Graphite	Graphite	Forced air
Brookhaven (Brookhaven National Laboratory)	Thermal, heterogeneous	Graphite	Graphite	Forced air
BEPO and GLEEP (Harwell, England)	Thermal, heterogeneous	Graphite	Graphite	Forced air
Production (Sellafield, England)	Thermal, heterogeneous	Graphite	Graphite	Forced air
Production (Hanford, Wash.)	Thermal, heterogeneous	Graphite	Graphite	Ordinary water
ZEEP (Chalk River, Canada)	Thermal, heterogeneous	Heavy water	Graphite	Ambient air
ZOE (Chatillon, France)	Thermal, heterogeneous	Heavy water	Graphite	Ambient air
CP-3 (Argonne)	Thermal, heterogeneous	Heavy water	Graphite	Circulation of heavy water
JEEP (Kjeller, Norway)	Thermal, heterogeneous	Heavy water	Graphite	Circulation of heavy water
Swedish (Stockholm, Sweden)	Thermal, heterogeneous	Heavy water	Graphite	Circulation of heavy water
P-2 (Saclay, France)	Thermal, heterogeneous	Heavy water	Graphite	Nitrogen or carbon dioxide
NRX (Chalk River, Canada)	Thermal, heterogeneous	Heavy water	Graphite	Ordinary water
Enriched Uranium Fuel				
CP-3′ (Argonne)	Thermal, heterogeneous	Heavy water	Graphite	Circulation of heavy water
CP-5 (Argonne)	Thermal, heterogeneous	Heavy water	Graphite	Circulation of heavy water
Materials Testing Reactor (Arco, Idaho)	Thermal, heterogeneous	Ordinary water	Beryllium and graphite	Circulation of water
Swimming Pool type (Oak Ridge and others under construction)	Thermal, heterogeneous	Ordinary water	Ordinary water (plus beryllium oxide or graphite in some cases)	Free convection (or circulation) of water
Water Boiler type (Los Alamos Scientific Laboratory, North Carolina State College, and others)	Thermal, homogeneous	Ordinary water	Graphite	Water through tubes
Homogeneous Reactor Experiment (Oak Ridge)	Thermal, homogeneous	Ordinary water	Heavy water	Circulation of fuel solution
Experimental Breeder Reactor (Arco, Idaho)	Fast	None	Uranium	NaK alloy
Plutonium Fuel				
Fast Reactor (Los Alamos); dismantled 1958.	Fast	None	Uranium	Mercury

pressure so as to permit operation at high temperatures, can be used as both coolant and moderator. Xenon poisoning presents a problem, although it can be overcome in a manner that is somewhat wasteful of neutrons. Reactors in which most of the fissions are caused by neutrons of intermediate energy have been proposed as a compromise between fast and thermal reactors.

1.176. The classification of reactors according to their neutron energy is more fundamental, but it is also convenient to describe reactors on the basis of the relative arrangement of fuel and moderator. In a *heterogeneous* reactor the fuel is distributed in a fairly definite geometrical pattern or lattice within the mass of moderator. Examples of this type are CP-1, CP-2, the X-10, Hanford, and Brookhaven reactors.

1.177. A *homogeneous* reactor, on the other hand, is one in which the fuel material is more or less evenly dispersed throughout the moderator. A uniform mixture of solid fuel and moderator, although not strictly homogeneous in the physical sense, would be regarded as a homogeneous reactor, since it would behave as such from the standpoint of neutron interaction. A slurry, consisting of a uniform suspension of an insoluble uranium or thorium compound in light or heavy water, would also behave as a homogeneous system. The various Water Boilers and the HRE (Homogeneous Reactor Experiment) at Oak Ridge, consisting of an enriched uranium salt as fuel dissolved in water as moderator, are examples of homogeneous reactors.

1.178. The HRE is a special form of homogeneous reactor in which the fuel-moderator mixture circulates out of the reactor, through a heat exchanger, and then back to the reactor. One purpose of the HRE was to test the practicability of such a circulating fuel system. This type of reactor has many advantages. The structural material is limited to the vessel and pipes containing the chain-reacting solution of an enriched uranium salt in ordinary (or heavy) water, thus keeping parasitic losses to a minimum. The problem of poisoning by xenon can be solved by continuous removal of its parent, iodine, from the system. Portions of the fuel solution can be bled off for the separation of fission products and extraction of fissionable material, e.g., plutonium, without interfering with the operation of the reactor. Finally, the metallurgical isolation of the fuel and the fabrication of special fuel elements, which are necessary for most other reactors, are obviated

Remote Control and Shielding

1.179. It should be noted, in this connection, that one of the factors to be considered in the design of a reactor is the matter of loading with fuel and unloading the spent material. The latter, as already mentioned, is highly radioactive and cannot be approached with any degree of safety. The spent fuel elements must, therefore, be discharged by some remote-handling device. They are then stored, preferably under a considerable depth of water, until the radioactivity has decayed sufficiently for the material to be conveyed, with appropriate precautions, to the processing plant for removal of fission products, etc., and for recovery of the fuel still remaining.

1.180. Reference must be made, finally, to one other essential aspect of reactor construction and design, namely, the shielding of operating personnel from neutrons and other harmful radiations. This does not present any great difficulty with stationary reactors. In a sense, all that is necessary is to surround the reactor, on all sides, by sufficient material, e.g., concrete, to reduce the radiations to essentially harmless proportions. The X-10 reactor at Oak Ridge, for example, has a shield of concrete with a thickness of 7 ft. For a mobile reactor, as might be used in a submarine or airplane, such a massive shield would be out of the question. The problem of radiation shielding in these cases requires special attention, so as to provide the maximum of protection for personnel with a minimum weight of material.

Conclusion

1.181. From the foregoing, somewhat general, description of nuclear reactors, it will be clear that the design of such reactors involves a close interrelationship of several sciences with some of the more common aspects of engineering. In the field of physics, for example, the various interactions of fast and slow neutrons with atomic nuclei are important since these determine the critical size of the reactor, as well as its time behavior. Radioactive decay and the rates of neutron capture reactions must be understood, because upon them depend the rates of accumulation of both desirable, e.g., plutonium-239 or uranium-233, and undesirable products, e.g., fission product poisons. The interaction of various radiations with matter must be studied so that the most effective methods of shielding may be developed. Because of the changes in physical properties produced in solids by exposure to high temperatures and intense nuclear radiations, the physics of the solid state acquires special significance.

1.182. The extraction of fertile and fissionable materials from their ores and their fabrication into suitable forms involve both chemistry and metallurgy. The separation of relatively small quantities of plutonium from the complex mixtures of fission products and unchanged uranium present in the spent fuel, and the recovery of the uranium, represent difficult problems in chemistry and chemical engineering. The difficulties are accentuated by the radioactivity of the materials and by the necessity for keeping losses to the absolute minimum. Further, the chemical effects of radiations, especially on solutions, must be taken in consideration. It is seen, therefore, that a knowledge and understanding of various aspects of chemistry and chemical engineering are essential for the proper design of a complete chain-reacting system.

1.183. Because operating personnel must be protected from the harmful consequences of exposure to excessive amounts of radiation, certain standards of permissible or tolerance dosages must be established. This involves a biological study of the effects of various radiations on living cells. The development of

methods for the protection of personnel and the measurement of radiation intensities and degrees of exposure have become a specialized field, called *health physics*, in which physics and biology are combined. In this connection, as well as in connection with the direct operation of the reactor and its associated equipment, the design of instruments for the detection and measurement of various nuclear radiations has become important.

1.184. In addition, there are a number of significant problems among the more conventional aspects of engineering which must be mentioned. Perhaps the most outstanding is that concerned with heat transfer, because the heat fluxes possible in a nuclear reactor are much higher than those usually encountered in power plants. The thermal stresses in some reactors, coupled with the effects of radiation on the physical properties of materials, necessitate special attention to mechanical design. Other mechanical problems of a somewhat different type arise from the requirements of remote control of various processes and the remote handling of radioactive substances. Finally, the design and operation of automatic control and safety systems, using servomechanisms, involve the application of established principles in electricity and electronics.

1.185. It will be apparent that, although many problems of nuclear reactor engineering are essentially those with which all engineers should be familiar to some extent, at least, the nuclear chain reaction introduces certain restrictions which must be taken into consideration in the solution of these problems. It is necessary, therefore, for the nuclear reactor engineer to become familiar with some of the fundamental aspects of physics, chemistry, and radiation biology, in order that he may apply his engineering knowledge to full advantage.

Symbols Used in Chapter I

A	mass number
amu	atomic mass unit (1.660×10^{-24} gram)
c	velocity of light (3×10^{10} cm/sec)
C	conversion ratio
e	base of natural logarithms
E	energy
ev	electron volt (1.60×10^{-12} erg)
G	breeding gain
k	infinite multiplication factor
k_{eff}	effective multiplication factor
k_{ex}	excess multiplication factor
l	neutron generation time (or lifetime)
L	neutrons lost by leakage and nonfission capture
m	mass
M	migration length
Mev	million electron volts (1.60×10^{-6} erg)
n	neutron density (neutrons/cm^3)

N	number of nuclei (usually per cm^3)
p	specific power (watts/gram or kilowatts/kilogram of fissionable material)
P	nonleakage probability
Q	10^{18} Btu
r	isotopic ratio (N^{235}/N^{238})
R	radius (or similar dimension)
t	time
T	reactor period
T_{K}	absolute temperature on Kelvin scale
T_{R}	absolute temperature on Rankine scale
v	velocity (cm/sec)
Z	atomic number
α	σ_c/σ_f (ratio of nonfission to fission in pure fissionable material)
β	fraction of fission neutrons delayed
η	average number of neutrons liberated per neutron absorbed in fuel
ν	average number of neutrons liberated per fission
σ	cross section
σ_c	capture (nonfission) cross section
σ_f	fission cross section

PROBLEMS

1. The unit electrical charge is 4.80×10^{-10} esu and 1 volt is 1/300 esu; verify the statement in the text that 1 Mev is equivalent to 1.60×10^{-6} erg. Note that charge (in esu) $\times$ potential (in esu) = energy (in ergs).

2. Check the values given in Table 1.6 for the energy liberated by the complete fission of 1 pound of fissionable material, starting from the basis that 200 Mev of energy are released per nucleus undergoing fission.

3. Determine the decrease of mass (in grams) accompanying the complete fission of 100 lb of uranium-235.

4. The heat of combustion of carbon is 7860 calories per gram. Calculate the decrease in mass (in grams) that would be associated with the combustion of 100 lb of carbon. (Compare the result with that of the preceding problem.)

5. An anthracite coal, consisting essentially of 80 per cent carbon and 20 per cent impurities, has a heat of combustion of 11,500 Btu/lb. Estimate the energy in electron volts produced by the combustion of 1 atom of carbon.

6. The mass of the hydrogen atom (H) is 1.00813, that of deuterium (D) is 2.01472, and that of helium (He) is 4.00388 amu. Ordinary water contains 1 atom of deuterium to 6500 atoms of ordinary hydrogen. Determine the energy release in Btu per pound of water if (1) the hydrogen atoms were converted into helium by the reaction $4\text{H} \rightarrow \text{He}$, (2) if the deuterium atoms only underwent the reaction $2\text{D} \rightarrow \text{He}$.

7. A nuclear central-station electrical power plant has a capacity of 60,000 kw. If the thermal efficiency for the conversion of fission energy into electrical power is 20 per cent, how many pounds of uranium-235 will be used up in a year of continuous operation? How many tons of natural uranium would be required to supply this amount of fissionable material?

8. A heterogeneous system containing 1 atom of natural uranium metal to 100 atoms of carbon (as graphite) has an infinite multiplication factor of 1.06. Taking the density

of uranium as 19 g/cm³ and that of graphite as 1.6 g/cm³, make a rough estimate of the critical weight (in tons) of uranium metal and graphite in a bare spherical reactor of the given composition. (The atomic weights of uranium and carbon may be rounded off to 238 and 12, respectively.)

9. The effective multiplication factor of a reactor, using natural uranium as fuel, is suddenly changed from the critical value to 1.0050. Determine the resulting reactor period and estimate how long it would take for the neutron density to increase by a factor of ten. How would these values compare with those to be expected from a further increase of 0.005 in the effective multiplication factor?

10. A homogeneous solution of a salt of natural uranium in heavy water has an infinite multiplication factor of 1.05; the migration length of thermal neutrons in the solution is 25 cm. Examine the prospects for a reactor, to produce plutonium and power, based on this system.

11. A source of 10-Mev neutrons is placed in a homogeneous mixture of natural uranium and graphite. Review the possible reactions which may occur.

12. What is the ratio of nonfission to fission capture of neutrons by plutonium for which breeding would just become possible, provided there were no loss of neutrons?

Chapter II *

NUCLEAR REACTIONS AND RADIATIONS

INTRODUCTION

2.1. The purpose of this chapter is to present such fundamental aspects of nuclear physics as are required for an understanding of the processes occurring in a nuclear reactor. These processes include spontaneous radioactive decay of uranium, thorium, plutonium, etc., as well as of the numerous products formed in fission. Consideration is given to the mechanism of nuclear reactions involving neutrons, notably radiative capture and fission, the latter process being treated in some detail. Equations are developed for the rates of nuclear reactions and it is shown how they may be combined to determine the concentrations of various isotopes that can be formed in a reactor. This information is important from the standpoint of production of fissionable material, on the one hand, and of poisoning by fission products, on the other hand.

2.2. Essentially all the processes occurring in a reactor, including radioactive decay and neutron reactions, are accompanied by the emission of nuclear radiations of different kinds. The interactions of these radiations with matter provide the basis for the methods used for their detection as well as for the development of suitable protective shielding. The following sections will deal with the general characteristics of nuclear reactions and radiations in so far as they are significant for reactor design and operation.

RADIOACTIVITY

Radioactive Isotopes

2.3. The naturally occurring forms of the great majority of elements are stable, but a few of high atomic weight, from polonium (atomic number 84) onward, e.g., radium (88), thorium (90), and uranium (92), consist entirely of unstable, radioactive isotopes. In addition, the elements thallium (81), lead (82), and bismuth (83) exist in nature largely as stable isotopes, but also to some extent as radioactive species. The unstable substances undergo spon-

* Reviewed by A. H. Snell.

taneous change, i.e., radioactive disintegration or radioactive decay, at definite rates. The decay is accompanied by the emission from the atomic nucleus of an electrically charged particle, either an *alpha particle*, i.e., a helium nucleus, or a *beta particle*, i.e., an electron.* Frequently the products of decay are themselves radioactive, expelling either an alpha or a beta particle. After a number of stages of disintegration, an atomic species with a stable nucleus is formed.

2.4. Besides the radioactive substances referred to above, all of which are found in nature, there have been produced in recent years a total of about 800 so-called "artificial" radioisotopes of all the known elements. These have been obtained either by bombardment of stable elements with charged particles in cyclotrons, etc., by the capture of neutrons (§ 1.58), or as a result of nuclear fission. A few of them expel alpha particles, but a large proportion, including most of the fission products, are negative-beta emitters. A number of artificial radioactive species emit positive beta particles, i.e., positrons or positively charged electrons, but such substances play essentially no part in nuclear reactors.

Rate of Radioactive Decay

2.5. For a given radioactive species, every nucleus has a definite probability of decaying in unit time; this decay probability has a constant value characteristic of the particular radioisotope.† It remains the same irrespective of the chemical or physical state of the element at all readily accessible temperatures and pressures. In a given specimen, the rate of decay at any instant is always directly proportional to the number of radioactive atoms of the isotope under consideration present at that instant. Thus, if N is the number of the particular radioactive atoms (or nuclei) present at any time t, the decay rate is given by

$$\frac{dN}{dt} = -\lambda N, \tag{2.5.1}$$

where λ, called the *decay constant* of the radioactive species, is a measure of its decay probability. Upon integration between any arbitrary zero time, when the number of radioactive nuclei of the specified kind present is N_0, and a time t later, when N of these nuclei remain, it is found that

$$N = N_0 e^{-\lambda t}. \tag{2.5.2}$$

Radioactive decay is seen to be an exponential process, the actual decay rate being determined by the decay constant λ and by the number of the particular nuclei present.

2.6. The reciprocal of the decay constant, represented by t_m, is called the

* Although a negatively charged beta particle and an electron are identical, electrons originating in atomic nuclei are generally referred to as beta particles.

† It now appears that in certain circumstances the decay probability may be changed very slightly, but this possibility may be ignored here.

mean life or average life of the radioactive species; thus,

$$t_m = \frac{1}{\lambda}. \tag{2.6.1}$$

It can be shown that the mean life is equal to the average life expectancy of the nuclei present at any time.

2.7. The most widely used method for representing the rate of radioactive decay is by means of the *half life*. It is defined as the time required for the number of radioactive nuclei of a given kind, or for their activity, to decay to half its initial value. Because of the exponential nature of the decay, this time is independent of the amount of the radioisotope present. Thus, if in equation (2.5.2), N is set equal to $\frac{1}{2}N_0$, the corresponding time $t_{\frac{1}{2}}$, which is the half life, is given by

$$e^{-\lambda t_{\frac{1}{2}}} = \tfrac{1}{2}$$

or

$$t_{\frac{1}{2}} = \frac{\ln 2}{\lambda} = \frac{0.6931}{\lambda}. \tag{2.7.1}$$

The half life is thus inversely proportional to the decay constant or, by equation (2.6.1), directly proportional to the mean life, i.e.,

$$t_{\frac{1}{2}} = 0.6931 t_m. \tag{2.7.2}$$

The half lives of known radioactive species range from a small fraction, e.g., about a millionth, of a second to billions of years.

2.8. Since the number of nuclei (or their activity) decays to half its initial value in a half-life period, the number (or activity) will fall to one fourth by the end of two periods, and so on (Fig. 2.1). In general, the fraction of the initial

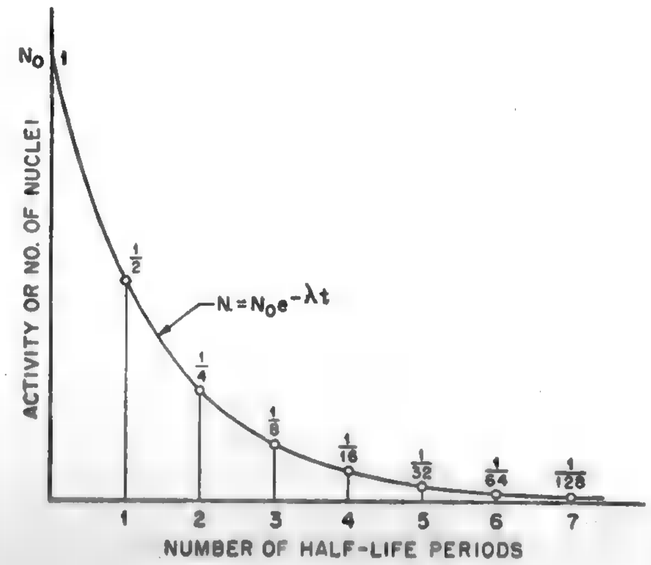


Fig. 2.1. Representation of exponential radioactive decay in terms of half-life periods

number of nuclei (or activity) remaining after n half-life periods is $(\frac{1}{2})^n$. After seven half-life periods, the amount of radioactive material has decreased to less than 1 per cent of its initial value.

2.9. The half lives of a number of substances of interest in the nuclear energy field are quoted in Table 2.1. On the left side are given the half lives of species

TABLE 2.1. CHARACTERISTICS OF RADIOACTIVE SPECIES

Naturally Occurring			Artificial		
Species	Activity	Half Life	Species	Activity	Half Life
Thorium-232	Alpha	1.39×10^{10} yr	Thorium-233	Beta	23 min
Uranium-238	Alpha	4.5×10^{9} yr	Protactinium-233	Beta	27.4 days
Uranium-235	Alpha	8.9×10^{8} yr	Uranium-233	Alpha	1.62×10^{5} yr
			Uranium-239	Beta	23 min
			Neptunium-239	Beta	2.3 days
			Plutonium-239	Alpha	2.41×10^{4} yr

which exist in nature, while on the right are those of artificial substances. Thus, thorium-233 is formed from thorium-232 by neutron capture (§ 1.61); it then decays to protactinium-233, which in turn yields uranium-233. Similarly, uranium-239 results when uranium-238 captures a neutron (§ 1.59); the successive decay products are then neptunium-239 and plutonium-239.

2.10. It is of interest to note that the three fissionable species, namely, uranium-233, uranium-235, and plutonium-239, and the two fertile substances thorium-232 and uranium-238, all of which are alpha emitters, have long half lives and so are comparatively stable. On the other hand, the substances produced when the fertile nuclei capture neutrons, i.e., thorium-233 and uranium-239, and their immediate decay products, i.e., protactinium-233 and neptunium-239, which are all beta emitters, have relatively short half lives. It means that these particular fertile and fissionable isotopes can be kept for many years without appreciable loss. On the other hand, since the intermediate stages following upon the capture of neutrons by fertile material have short lives, they will decay almost completely within a few days to form fissionable species.

Radioactive Equilibrium

2.11. In a series of decay stages, such as those which follow nonfission neutron capture by thorium-232 or uranium-238, or as represented by the members of the natural radioactive series, or as exhibited by many fission products, each radioactive member of the series decays in accordance with equation (2.5.1), with its own specific value for the decay constant. Such a series may be represented by

$$A \xrightarrow{\lambda_A} B \xrightarrow{\lambda_B} C \xrightarrow{\lambda_C} D \xrightarrow{\lambda_D} \cdots \cdots \rightarrow X,$$

where A may be the parent of a natural radioactive series, or the species formed when a neutron is captured, e.g., thorium-233 or uranium-239, or it may be a radioactive fission product; the stable end-product of the series is represented by X. Consider any member of the series, other than the first or last, such as B, for example. While the rate of decay is given by equation (2.5.1), it must be remembered that, at the same time, B is being formed by decay of A; hence, at any specified time, the net rate of change of B with time is given by

$$\frac{dN_B}{dt} = \lambda_A N_A - \lambda_B N_B, \tag{2.11.1}$$

the first term on the right representing the rate of formation of B, by decay of A, and the second term the rate of decay of B. A similar relationship applies to other members of the series.

2.12. If N_A is constant, as it is virtually in the natural radioactive series because of the very long half life of the parent, then a condition will sooner or later be reached when $\lambda_A N_A$ is equal to $\lambda_B N_B$, so that, by equation (2.11.1), the net rate of change of B is zero. In other words, A is decaying to form B at the same rate as B is decaying to form C, so that the amount of B in the system remains constant. This is called a state of *radioactive equilibrium*. In due time, all the members of a radioactive series, except the last, will be in radioactive equilibrium with one another, provided the amount of the parent remains essentially unchanged.

2.13. Although in a reactor the situation is somewhat different, because the parents are formed by fission or by nonfission neutron capture, it will be shown later (§ 2.186, *et seq.*) that, provided the power (or neutron density) remains constant, a similar condition of equilibrium is attained. When the reactor is shut down or the radioactive material is removed, e.g., as spent fuel, the equilibrium condition is disturbed. The rate of change in the amounts of the various members of the series can then be determined by solving a set of differential equations of the form of equation (2.11.1).

Radioactivity Units: The Curie

2.14. The *curie*, which was based on the estimated activity of a gram of radium, is now defined as the quantity of any radioactive species decaying at a rate of 3.7×10^{10} disintegrations per second (dis/sec). A sample of radioactive material is said to have an activity of 1 curie when it disintegrates at this rate. Subsidiary units, for smaller quantities of active material, are the *millicurie* (1 mc), which is a one-thousandth part of a curie, i.e., 3.7×10^{7} dis/sec, and the *microcurie* (1 μc), a one-millionth of a curie, i.e., 3.7×10^{4} dis/sec. For large amounts of radioactive substances two other units are often employed. These are the *kilocurie*, which is a thousand curies, and the *megacurie*, i.e., a million curies.

2.15. It was shown above that a specimen containing N atoms (or nuclei) of

a given radioisotope decays at the rate λN, and since λ is equal to $0.693/t_{\frac{1}{2}}$, where $t_{\frac{1}{2}}$ is the half life, it follows that

$$\text{Rate of decay} = \frac{0.693N}{t_{\frac{1}{2}}} \text{ dis/sec}, \tag{2.15.1}$$

if $t_{\frac{1}{2}}$ is in seconds. The number of individual atoms (or nuclei) present in 1 gram atom, i.e., the atomic weight in grams, is the Avogadro number, 6.02×10^{23} (§ 1.87). Hence, if g grams is the weight of the radioisotope, of atomic weight A, present in the specimen, equation (2.15.1) leads to

$$\text{Rate of decay} = \frac{(0.693)(6.02 \times 10^{23})g}{At_{\frac{1}{2}}} \text{ dis/sec}. \tag{2.15.2}$$

Upon introducing the definition of the curie, i.e., 3.7×10^{10} dis/sec, it is found that

$$\text{Number of curies} = \frac{1.13 \times 10^{13}\, g}{At_{\frac{1}{2}}}. \tag{2.15.3}$$

The mass of the substance having an activity of 1 curie, i.e., g/curies, is then

$$\text{Mass per curie} = \frac{At_{\frac{1}{2}}}{1.13 \times 10^{13}} \text{ gram}. \tag{2.15.4}$$

The mass of a particular radioisotope having an activity of 1 curie thus depends upon both its atomic weight and its radioactive half life. For an isotope that decays slowly, i.e., one with a long half life, the mass per curie will, apart from the effect of atomic weight, be greater than for a rapidly decaying species.

2.16. Some confusion and error have arisen in the determination of the activity of a material in curies because of a failure to distinguish between the rate of disintegration and the rate of emission of particles. If a certain radioisotope emits a single type of radiation, e.g., beta particles only, then the rates of decay and of particle emission are the same. However, there are cases in which two particles are formed, e.g., a beta particle and a gamma-ray photon (see § 2.21), and not necessarily in equal numbers. In such instances the total rate of particle emission is not a measure of the activity in curies. The latter is defined in terms of the actual rate of disintegration, and data based on particle emission may require correction for various reasons.

2.17. Before the discovery of nuclear fission, a 1-curie amount of radioactive material was considered to be very highly active. The operation of nuclear reactors has, however, led to the formation of relatively large quantities of intensely radioactive fission products. Consequently, activities of the order of thousands or even millions of curies, i.e., kilocuries or megacuries, are now not unusual for the fuel elements immediately after removal from a reactor (see Chapter VII).

2.18. It may be mentioned here that a unit of a different type, which is related to the effect produced by (or energy absorbed from) the radiation emitted by a radioactive material, is used in health physics work. This unit is called the "roentgen": it will be considered more fully in Chapter IX.

Gamma Rays and X-rays

2.19. In many instances, when a nucleus suffers radioactive decay, the product (or daughter) nucleus is not in its lowest energy state or ground state. In other words, the product nucleus is in an excited state having energy in excess of the ground state. Within a very short time, perhaps 10^{-14} sec, of its formation, the excited nucleus emits the excess (or excitation) energy in the form of electromagnetic radiation, called *gamma rays* (Fig. 2.2). These rays are similar in character to X-rays; they are highly penetrating and have short wave lengths, usually in the range of 10^{-9} to 10^{-11} cm.

2.20. Although the term gamma rays was originally used, as indicated above, to describe the electromagnetic radiations which frequently accompany radioactive decay, the definition has now been extended to include all such radiations of nuclear origin. Examples are the radiations accompanying fission and neutron reactions of the radiative capture type (§ 1.57), as well as those associated with inelastic scattering processes (§ 1.51). In each case, the rays are emitted when a nucleus undergoes transition from a higher-energy to a lower-energy state.

2.21. According to the quantum theory of radiation, the energy of the gamma rays (and of other electromagnetic radiations) is emitted as *photons;* these may be regarded as "atoms" or "particles" of the radiation concerned. The energy of the photon is equal to the difference in energy between the two energy states involved in the transition, i.e., to the energy E in Fig. 2.2.

2.22. If the energy transition takes place directly from the excited state to the ground state, one photon will carry the whole of the excitation energy. In some cases, however, the transition occurs in two (or more) stages, e.g., from a

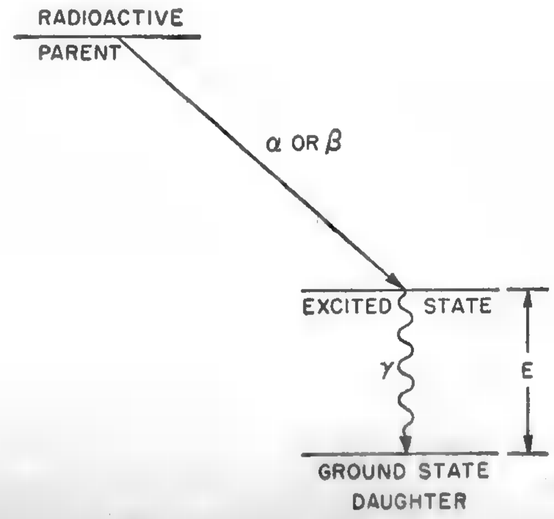


Fig. 2.2. Emission of gamma rays in radioactive change

higher to a lower excited state and then to the ground state. The total excitation energy is then divided among two (or more) photons, the energy of each being equal to the energy change in the corresponding transition (Fig. 2.3).

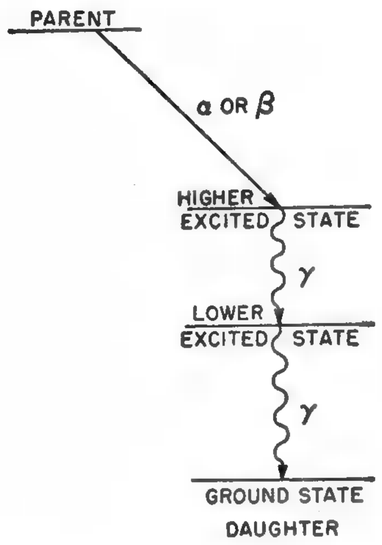


FIG. 2.3. Emission of two gamma-ray photons in radioactive change

2.23. It has become the common practice to describe gamma radiation in terms of its photon energy. For example, the expression "1-Mev gamma rays" refers to radiation with photons carrying 1 Mev of energy. It may be noted that nuclear excitation energies are generally of the order of Mev's, usually ranging from one tenth to ten million electron volts. The energies of gamma rays are consequently of this order of magnitude.*

2.24. Apart from the fact that X-rays frequently have lower energies, the essential difference between gamma rays and X-rays is that the latter are produced outside the atomic nucleus. The characteristic X-rays, which, as their name implies, have definite energies (and wave lengths) characteristic of the particular element concerned, result from transitions between electronic energy levels of the atom. These radiations are, however, of little significance for the present purpose. Of greater interest are the continuous X-rays, frequently called *bremsstrahlung*, i.e., braking (or slowing down) radiations, covering a considerable range of energies. These are produced when electrons (or beta particles) of high speed are slowed down as a result of interaction with matter.

2.25. As a general rule, the fraction of the kinetic energy of the electrons converted into radiation in this manner increases with the energy of the electron and with the atomic number of the material in which it is slowed down. The energy of the resulting X-rays covers a large range; the maximum is close to that of the electrons (or beta particles), but the average energy is much less. When electrons of energies of 1 Mev or more interact with elements of high atomic number, e.g., lead, some of the resulting bremsstrahlung, although originating outside the nucleus, are indistinguishable in their behavior from gamma rays arising from nuclear transitions.

INTERACTION OF RADIATIONS WITH MATTER

Ionizing Radiations

2.26. In its passage through matter, a fast-moving charged particle, such as an alpha or a beta particle, will occasionally approach close enough to an atom

* The wave length of gamma rays can be related to the photon energy by means of the quantum theory; thus, wave length (in cm) $= 1.24 \times 10^{-10}/E$ (in Mev).

(or a molecule) for the electrical interaction to be sufficient to remove an external (or orbital) electron from the atom. The residue of the atom, after removal of the electron, is a positively charged ion, and the system of the separate electron and ion so formed is called an *ion-pair*.* Electrically charged particles are thus able to cause ionization, and, as they traverse matter, they leave behind a number of ion-pairs in their path. For this reason, alpha and beta particles are frequently referred to as *ionizing radiations*.

2.27. The intensity of the ionization produced by a moving charged particle is expressed by the *specific ionization;* this is the number of ion-pairs formed per cm of path in a given material. For charged particles of the same mass, the specific ionization increases with the charge. For particles of the same energy, those of higher mass produce a higher specific ionization. Since they move more slowly, the heavier particles (of given energy) spend more time in the vicinity of a given atom or molecule of the material through which they pass, thus increasing the probability of ionization.

2.28. The specific ionization due to an alpha particle, mass about 4 amu (§ 1.43), is thus considerably greater than that caused by a beta particle, mass 0.00055 amu, of the same energy. In their passage through air at atmospheric pressure, alpha particles produce approximately 50,000 to 100,000 ion-pairs per cm, whereas beta particles of similar energy form about 30 to 300 ion-pairs/cm.

2.29. The energy required to produce an ion-pair varies from one medium to another. Consequently the specific ionization produced by a charged particle depends on the nature of the medium through which it passes, as well as on the charge, energy, and mass of the particle. In its passage through air, a charged particle loses, on the average, about 32.5 ev of energy for every ion-pair produced. This energy varies somewhat with the nature of the particle, but the figure quoted may be accepted for most purposes. It is seen, therefore, that an alpha particle having an initial energy of 5 Mev will be capable of forming something like 1.5×10^5 ion-pairs in air. Since the specific ionization is roughly 5×10^4 ion-pairs/cm, the ionization will occur over a path length (or *range*) of about 4 cm. The effective range for a beta particle of the same energy will be nearly a thousand times greater.

2.30. Gamma rays and X-rays do not carry an electric charge and so are not ionizing radiations. Nevertheless, as will be seen below, in their passage through matter these rays eject electrons and the latter, being electrically charged, are able to produce ionization. Gamma rays and X-rays thus cause ionization indirectly; they can also cause electronic excitation directly or indirectly. Neutrons, which are also electrically neutral, behave somewhat similarly, although a different mechanism is involved. The direct ionization (and excitation) caused by alpha and beta particles and the indirect ionization (and

* In some interactions the energy transferred from the radiation to the atom or molecule may be insufficient to cause ionization, i.e., removal of an electron, but may be sufficient to raise the electron to a higher energy level, so that the atom or molecule is in an excited electronic state.

excitation) due to gamma rays, X-rays, and neutrons are important in connection with methods of measuring or counting these nuclear radiations and also with their effects on living tissue.

2.31. The majority of the separated positive ions and electrons formed, directly and indirectly, when nuclear radiations traverse matter, will sooner or later reunite to produce neutral atoms or molecules. This process is accompanied by the liberation of energy in the form of heat. Hence essentially all the energy of nuclear radiations is ultimately degraded into heat, as stated in Chapter I. Some of the charged particles may become involved in chemical action; in this event, a portion of the energy of the radiation is converted into chemical energy.

Absorption of Alpha Particles

2.32. If the specific ionization caused in air by alpha particles from a given source is measured at various distances from the source, a curve of characteristic shape, called a *Bragg curve*, is obtained. A typical Bragg curve is shown in Fig. 2.4. It is seen that the specific ionization increases with increasing distance from the source, at first slowly and then rapidly; after passing through a maximum, it drops sharply to zero. The increasing specific ionization is accounted for by the fact that, as the alpha particle traverses its path, producing ion-pairs as it proceeds, its energy and, consequently, its speed decrease steadily. The specific ionization, which increases with decreasing speed, as stated above, thus increases with distance from the source. Near the end of its path, when the alpha particle is moving relatively slowly, the specific ionization reaches a maximum.

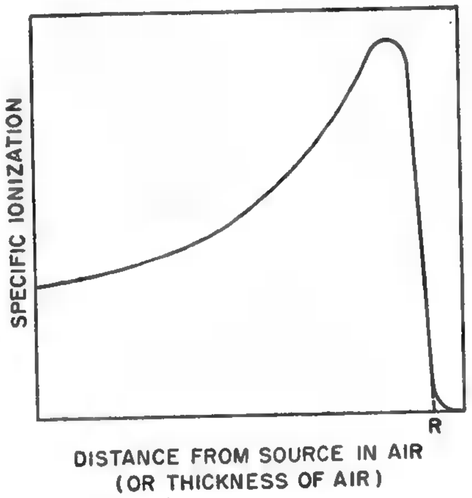


Fig. 2.4. Specific ionization and range of alpha particles

2.33. Beyond a certain point, the energy of the alpha particle has become so low that it captures one and then two electrons, being thus converted into a neutral helium atom no longer capable of producing ionization. This explains the decrease in the specific ionization shown in Fig. 2.4. The fact that this decrease is relatively sudden, and not gradual, is of interest; it implies that all the alpha particles from a given source have practically the same energy, and all cease to produce ionization after having traveled the same distance. Strictly speaking, if all the alpha particles behaved exactly alike, the end portion of the curve would be vertical, but minor differences account for the slight slope. The extrapolated distance, represented by R, at which alpha particles from the given source effectively cease to produce ionization, is called the *range* of the particles.

2.34. The range of alpha particles in air (or in other material) depends on the source; for particles of radioactive origin, it varies from 2.5 cm (1.0 in.) for thorium-232 to 8.6 cm (3.4 in.) for thorium C′. There is an inverse relationship between the half life of the radioisotope and the energy, and hence the range, of the alpha particles. Thus, the shorter the half life, the greater the alpha-particle range. Some of the data for alpha-particle emitters of interest in nuclear reactors are given in Table 2.2. Uranium-235 produces two groups of alpha particles with slightly different energies and ranges.

Table 2.2. Half Lives, Energies, and Ranges in Air

Radioelement	Half Life (yr)	Energy (Mev)	Range in Air	
			(cm)	(in.)
Thorium-232	1.39×10^{10}	4.0	2.5	1.0
Uranium-238	4.5×10^{9}	4.2	2.7	1.06
Uranium-235	8.9×10^{8}	4.4 and 4.6	2.9 and 3.1	1.15 to 1.2
Uranium-233	1.62×10^{5}	4.8	3.3	1.3
Plutonium-239	2.41×10^{4}	5.1	3.6	1.4

2.35. In absorbing materials more dense than air the specific ionization is, in general, greater because of the greater concentration of atoms, and hence the range of alpha particles of given energy is less than in air. The relative *stopping power* of an absorber is defined by

$$\text{Relative stopping power} = \frac{\text{Range of alpha particle in air}}{\text{Range of alpha particle in material}}. \qquad (2.35.1)$$

It is essentially independent of the alpha-particle energy, provided, of course, that the ranges refer to particles from the same source.

2.36. The relative stopping power of water is about 1000, for aluminum 1600, and for lead it is over 5000. Paper and animal tissue have roughly the same stopping power as water, i.e., around 1000. If an alpha particle has a range of, say, 2 in. in air, which is relatively high, the range in paper or living tissue would be about 0.002 in., i.e., 2 mils. Thus, an ordinary sheet of paper, having a thickness of roughly 4 mils, would stop essentially all alpha particles of radioactive origin.

2.37. Another method for representing the absorbing effect of a medium is by means of the *thickness density*, frequently referred to, in brief, as the "thickness." It is the product of the linear range of the given particles in the medium and the density of the medium; thus, if the range is R cm and the density is ρ g/cm^3, then

$$\text{Thickness density} = R \times \rho \text{ g/cm}^2 = 1000R \times \rho \text{ mg/cm}^2, \quad (2.37.1)$$

according as the value is expressed in g/cm² or mg/cm². The latter is generally used because the numbers are then of a reasonable magnitude. Physically, the thickness density is the mass per unit area of absorbing medium required to stop (or absorb) the given alpha particles, i.e., having a linear thickness equal to the range of the particles.

Absorption of Beta Particles

2.38. The passage of beta particles through matter has some features in common with the behavior of alpha particles, e.g., production of ion-pairs at the rate of about 32.5 ev/ion-pair in air, but there are some important differences. As stated earlier, the smaller mass of the beta particle means that the specific ionization is less than that due to an alpha particle of the same energy. Further, all the alpha particles from a given source have essentially the same energy, or they fall into two or three groups of definite energy. Beta particles, on the other hand, have a continuous distribution of energies, i.e., a continuous energy spectrum, up to a fairly definite maximum for each particular source (Fig. 2.5). This maximum energy is believed to be equal to the total energy of the radioactive transition, and it has been suggested that the difference between the maximum and the actual energy of the beta particle is carried off by an electrically neutral particle of negligible mass, called a *neutrino* (cf. § 1.58, footnote).*

2.39. Because of their relatively large mass, alpha particles do not, on the whole, undergo any marked change of direction in their passage through matter. In other words, the majority travel in straight lines, thus leading to a fairly definite range for a given energy. Beta particles, however, are subject to considerable scattering, with frequent changes in direction as a result of collisions with atomic nuclei and electrons. Consequently, beta particles which have passed through the same thickness of a given absorber may come out in widely different directions, so that they will actually have traversed paths of different lengths in the material.

2.40. The combined effect of the continuous energy spectrum of beta particles and their scattering means that these particles do not have a definite range, as do alpha particles from a given source. However, due to a fortuitous combination of circumstances, which are too complex for complete theoretical analysis, it is found experimentally that the ionization caused by beta radiation from a given source falls off in a roughly exponential manner with distance. The general form of the plot of the logarithm of the ionization produced against the thickness of the absorbing material is shown in Fig. 2.6; except for small or large thicknesses, the curve is approximately linear. At large absorber thick-

*The average energy of the beta particles from a given source is about one third of the maximum value.

nesses the curve becomes almost horizontal, indicating a more or less constant ionization. This "tail" of the curve is due to the presence of the highly penetrating bremsstrahlung (§ 2.24) resulting from the deceleration of the fast-moving beta particles in their passage through the absorbing medium.

2.41. The rate of energy loss as bremsstrahlung per unit path length by beta particles (or electrons) is approximately proportional to the square of the atomic number of the absorber and to the energy of the particles. The association of bremsstrahlung with the passage of beta particles of given energy through matter is thus most marked with elements of high atomic number. The radiations cover a range of energies up to a maximum equal to the initial (maximum) energy of the beta particles.

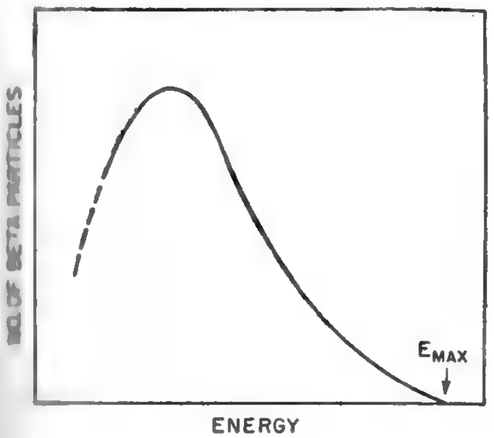


Fig. 2.5. Energy spectrum of beta particles

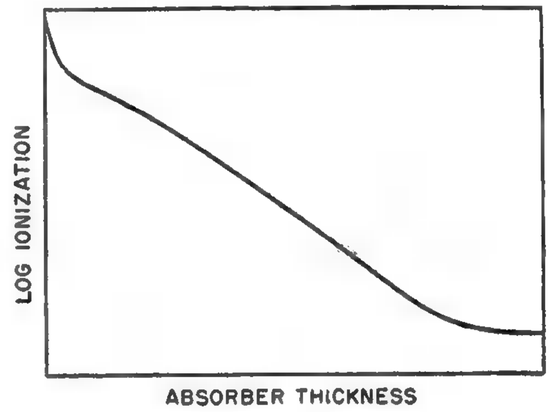


Fig. 2.6. Absorption of beta particles

2.42. If gamma rays are emitted from the beta-particle source they will, of course, also contribute to the "tail" in Fig. 2.6. After passage through considerable thicknesses of material, both gamma radiation and bremsstrahlung will be absorbed.

2.43. Although the beta particles do not have a definite range, in the same sense as do alpha particles, it is nevertheless possible to specify a more or less definite thickness of absorber which will reduce almost to zero the ionization due

TABLE 2.3. APPROXIMATE RANGES OF BETA PARTICLES IN AIR

Energy	*Range*	
(Mev)	(meters)	(ft)
0.1	0.11	0.36
0.5	1.5	4.9
1.0	3.7	12
2.0	8.5	28
3.0	13	43

to beta particles of given energy, apart from the effect of bremsstrahlung. The approximate ranges in air of beta particles of various maximum energies are given in Table 2.3. It may be noted that the average maximum energy of the beta particles from fission products is about 1.2 Mev; the absolute maximum energy probably does not exceed 3 Mev and may be appreciably less.

2.44. As is the case with alpha particles, the approximate range of beta particles in an absorbing material is frequently expressed in terms of its thickness density in grams per cm² as defined by equation (2.37.1). The values for aluminum as absorber have been determined experimentally for beta particles

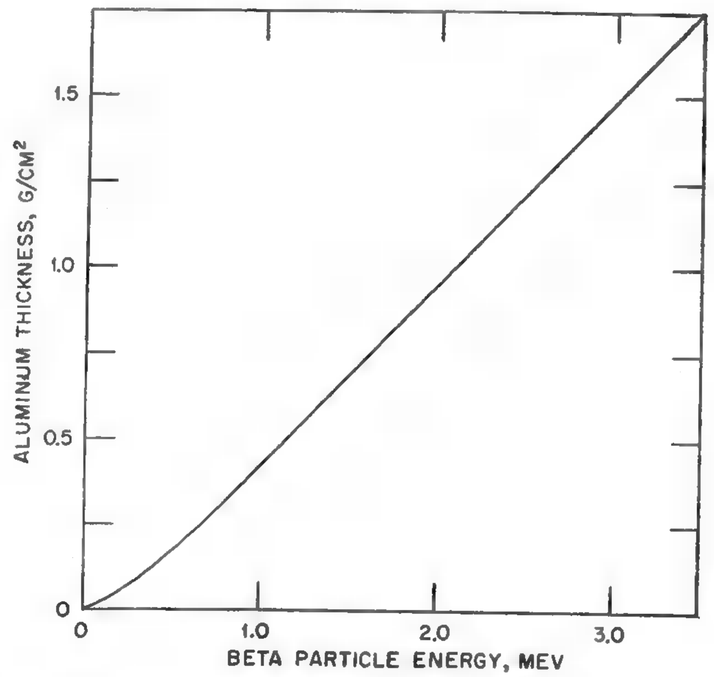


FIG. 2.7. Aluminum thickness density (g/cm²) as function of beta-particle energy

from various sources, and the results fall on or very close to the curve shown in Fig. 2.7; the ordinates are the thickness densities of aluminum in grams per cm² required to absorb beta particles with maximum energies indicated by the abscissae. Within the range of about 0.8 to 3 Mev, the relationship between the absorption thickness density ($R \times \rho$) of aluminum and the maximum energy (E_m) of the beta particles from the given source is linear and may be represented by the expression

$$R \times \rho \text{ (g/cm}^2\text{)} = 0.54E_m \text{ (Mev)} - 0.15. \qquad (2.44.1)$$

2.45. In the absence of other data, it may be supposed, as a first approximation, that the absorption thickness density is independent of the nature of the absorbing material; this is based on the assumption that the linear range of the

beta particles in any medium is inversely proportional to the density. In this event, the values of $R \times \rho$ given by equation (2.44.1) would be the same for all absorbers. The (approximate) linear range in any material can then be obtained if the density is known.

Example Estimate the maximum range in concrete of density 2.8 g/cm³ of beta particles from fission products.

As stated in § 2.43, the energy of the beta particles from fission products does not exceed 3 Mev; hence, from equation (2.44.1), the corresponding maximum value of $R \times \rho$ is 1.47 g/cm². Since ρ is given as 2.8 g/cm³, it follows that

$$R = 1.47/2.8 = 0.53 \text{ cm}.$$

The maximum range is thus about 0.53 cm, or 0.21 in., in concrete.

2.46. Positive beta particles, i.e., positrons, behave like negative beta particles in their interaction with matter. There is, however, another factor to be considered in the former case. Within a very short time of its liberation, a positron is likely to combine with an electron, of which large numbers are always present in matter as the outer electrons of atoms. The positively charged positron and negatively charged electron neutralize one another; the particles are thereby annihilated and energy is liberated in the form of radiation, called *annihilation radiation*. The total mass of a positron and an electron is 0.00110 amu and, by equation (1.76.2), this is equivalent to 1.02 Mev. In order that momentum may be conserved this energy is usually divided equally between two photons moving in opposite directions. The energy of the annihilation radiation, which has properties similar to gamma radiation, is thus mainly 0.51 Mev, although there may be a small proportion of 1.02-Mev energy.

Interaction of Gamma Rays with Matter

2.47. Although X-rays, bremsstrahlung, and annihilation radiation are not strictly gamma rays, since they do not arise from nuclear transitions, they are essentially identical with gamma rays in their fundamental nature. As far as their interaction with matter is concerned, the only differences that may arise are the result of the higher energies, in general, of the gamma radiations. The discussion in the following paragraphs may, therefore, be taken as applicable to all electromagnetic radiations of high energy, e.g., 0.01 to 100 Mev.

2.48. There are several ways in which gamma rays interact with an absorbing material; three, namely, the photoelectric effect, the Compton effect, and pair production, are important and will be considered here in some detail. Certain other reactions due to gamma rays will be mentioned elsewhere (§ 2.74).

Photoelectric Effect

2.49. In the photoelectric effect, a gamma-ray photon, with energy greater than the binding energy of an orbital electron in an atom, interacts with the latter in such a way that the whole of the gamma-ray energy is transferred to an

electron which is consequently ejected from the atom. If E is the energy of this gamma-ray photon and B is the binding energy of the electron in the atom, the difference, i.e., $E - B$, is carried off as kinetic energy by the ejected electron. The photoelectron, as it is called, behaves like a beta particle of the same energy in its passage through matter. For gamma rays of high energy, the photoelectrons are mainly expelled in the forward direction, i.e., in the same direction as the incident gamma rays, but for low-energy rays, the emission is largely in the direction at right angles.

2.50. The extent of the photoelectric interaction depends on both the energy, E, of the gamma radiation and the atomic number, Z, of the absorbing material. As a rough approximation,

$$\text{Probability of photoelectric interaction} \approx \text{const.} \times \frac{Z^n}{E^3},$$

where n varies from 3, for gamma rays of low energy, to 5, for high-energy rays. It is apparent, therefore, that the photoelectric effect increases with increasing atomic number of the absorber and with decreasing energy of the gamma rays. In actual practice, it is found that photoelectric absorption of gamma radiation is important only for energies less than about 1 Mev, and then only for absorbers of high atomic number (cf. Figs. 2.10 and 2.11 below).

2.51. Following the expulsion of the photoelectron, another electron, from an outer orbit, takes its place in the atom; this transition is accompanied by the emission of characteristic X-rays. These are of low energy, as compared with the original gamma radiation. The expulsion of the X-ray photon frequently causes the ejection of an outer electron, called an Auger electron, in a type of photoelectric (Auger) effect; the photon thereby loses all of its energy. The Auger electron will escape from the atom and will dissipate its energy as a result of interactions similar to those experienced by a beta particle. It is evident, from the foregoing discussion, that the photoelectric effect leads to the virtually complete absorption of the gamma-ray photon; it may be replaced, to some extent, by X-ray photons of low energy, but few of these escape as such from the absorbing material.

Compton Effect

2.52. In this process a gamma-ray photon makes an elastic (or "billiard-ball") collision with an outer electron of an atom of the absorbing material. Such an electron is loosely bound so that it behaves as if it were completely free. In the collision both momentum and energy are conserved, and part of the energy of the incident photon is transferred to the electron; at the same time the photon is deflected, i.e., scattered, from its initial path (Fig. 2.8). The relation between the energy E of the incident photon, E' of the scattered photon, both in Mev, and the scattering angle θ, is given by*

* The quantity 0.51 Mev, which appears in this equation, is m_0c^2, where m_0 is the mass of the electron at rest, and c is the velocity of light. It is the energy equivalent, according to the Einstein equation, of the rest mass of the electron (cf. ¶ 2.46).

$$E' = \frac{0.51}{1 - \cos\theta + 0.51/E}. \tag{2.52.1}$$

2.53. If the scattering angle is small, $\cos\theta \approx 1$, and then E' is approximately equal to E. This means that the scattered photons of higher energy, close to the energy of the incident photons, proceed in a nearly forward direction. On the other hand, for $\theta = 90°$, $\cos\theta = 0$, and then

$$E' = \frac{0.51E}{E + 0.51} < 0.51 \text{ Mev}.$$

Consequently a photon scattered at right angles cannot have energy greater than 0.51 Mev.

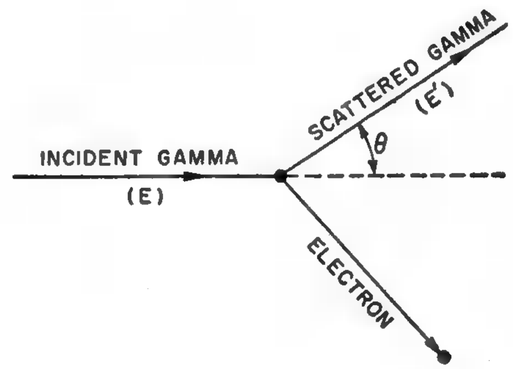


Fig. 2.8. Compton scattering of gamma-ray photon

2.54. The fraction of the initial energy carried by the scattered photon for different scattering angles is derived from equation (2.52.1) as

$$\frac{E'}{E} = \frac{0.51}{E(1 - \cos\theta) + 0.51}.$$

For a specified value of θ this fraction decreases with increasing energy of the incident photon. In other words, for a given scattering angle, the greater the energy of the incident photon, the smaller the fraction remaining on the scattered photon, and the larger the fraction (and amount) of energy lost by the gamma ray in the Compton interaction.

2.55. Since the Compton effect involves interaction between a photon and an electron, its magnitude is dependent on the number of orbital electrons in the atom of the absorber; this is the same as the atomic number. The Compton interaction is thus directly proportional to the atomic number of the absorber, so that, like the photoelectric effect, it is more significant for materials of high atomic number. The energy dependence of the Compton effect is given by the Klein-Nishina formula.* This is somewhat too complicated to reproduce here, but it can be stated that the Compton interaction decreases monotonically with increasing energy of the gamma radiation (cf. Figs. 2.10 and 2.11). As a very rough approximation, it is possible to write

$$\text{Probability of Compton interaction} \approx \text{const.} \times \frac{Z}{E}.$$

2.56. There is a significant difference between the photoelectric and Compton effects to which attention must be called. The photoelectric effect is a true absorption process; that is to say, the photon is absorbed, as stated above. In

* O. Klein and Y. Nishina, *Z. Physik*, **52**, 853 (1929); see also W. Heitler, "The Quantum Theory of Radiation," Oxford University Press, 3rd ed., 1954, p. 215, *et seq.*; D. Halliday, "Introductory Nuclear Physics," John Wiley and Sons, Inc., 1950, p. 152.

the Compton process, however, there is merely a decrease in the photon energy, the extent of this decrease being greater the larger both the initial energy and the scattering angle. A photon which has been scattered in a Compton interaction will thus still be present in the medium, although it will have a somewhat lower energy and may be moving in a different direction. Such a photon may suffer several Compton collisions in a sufficiently thick absorbing medium. The photon is then said to undergo multiple scattering. If it does not escape, the scattered photon will ultimately be absorbed as a result of photoelectric interaction which becomes increasingly probable as the energy decreases (§ 2.50).

Pair Production

2.57. When a gamma-ray photon with energy in excess of 1.02 Mev passes near the nucleus (and to some extent the outer electrons) of an atom, the photon can be annihilated in the strong electrical field with the formation of an electron-positron pair. Since the energy equivalent of the total mass of an electron and a positron is 1.02 Mev (§ 2.46), this is the minimum energy necessary for the production of the pair of particles. Any energy of the gamma-ray photon in excess of 1.02 Mev appears mainly as kinetic energy of the electron and positron, with a small fraction transferred to the atomic nucleus. The particles produced tend to travel in the forward direction, the effect becoming more evident with increasing gamma-ray energy.

2.58. The extent of pair production by gamma radiation of energy E (in Mev) is related to the atomic number Z of the absorber by

$$\text{Probability of pair production} \approx \text{const.} \times Z^2(E - 1.02),$$

so that it increases with the atomic number of the absorbing material and with increasing photon energy in excess of 1.02 Mev. Actually this expression allows only for pair formation in the electrical field of the nucleus, but in addition the process occurs, to some extent, in the fields of the orbital electrons. This contribution also increases with the atomic number. Since both the photoelectric and Compton effects decrease with increasing gamma-ray energy, whereas pair production increases, it is evident that the latter process will become of greater importance at high energies. For absorbers of high atomic number it becomes the dominant type of interaction for gamma rays with energies in excess of about 5 Mev.

2.59. As in the photoelectric effect, pair production results in the absorption of the gamma-ray photon. It is true that some of the electrons and positrons formed will neutralize one another and produce annihilation radiation, consisting, in general, of two 0.51-Mev photons (§ 2.46). However, because of the relatively low energy of this radiation, and the fact that it has an isotropic distribution, i.e., it is spread uniformly in all directions, so that the proportion continuing in the forward direction is small, it may be assumed for many practical purposes, e.g., in shielding, that the photon is completely absorbed in pair production.

Absorption of Gamma Rays

2.60. In their passage through matter the gamma-ray photons are absorbed so that the intensity falls off in an exponential manner. This arises from the fact that the extent of absorption in a small thickness dx of matter, at any point in the medium, is proportional to the radiation intensity at that point and to the thickness traversed, i.e.,

$$dI = -\mu I\, dx \quad \text{or} \quad \frac{dI}{I} = -\mu\, dx, \tag{2.60.1}$$

where I is the intensity, expressed as photons (or Mev) per cm^2 per sec, and the proportionality constant, μ, usually given in cm^{-1} units, is called the *linear absorption coefficient* of the absorber for the given radiation. If a collimated beam of monoenergetic gamma rays of intensity I_0 passes through a thickness x cm of absorber, the intensity I_x of the emergent beam is obtained by integration of equation (2.60.1); the result is

$$I_x = I_0 e^{-\mu x} \tag{2.60.2}$$

or

$$\log \frac{I_x}{I_0} = -0.4343\mu x. \tag{2.60.3}$$

The general character of the variation of the radiation intensity with thickness of the absorber is shown in Fig. 2.9, which is a representation of equation (2.60.3). It should be mentioned that this expression is particularly applicable to small or moderate thicknesses of absorber, so that photons which have suffered Compton scattering are essentially lost from the collimated beam.

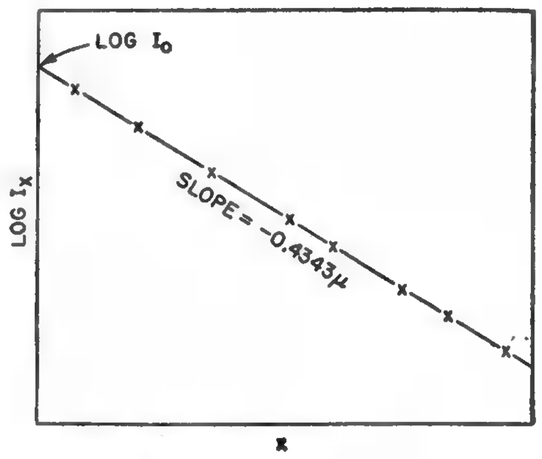


Fig. 2.9. Variation of gamma-ray intensity with absorber thickness

2.61. One of the consequences of the exponential absorption of gamma rays is that, although the *amount* of radiation absorbed by a specified thickness of material is proportional to the intensity, the *fraction* absorbed (or emerging) is independent of this intensity. For some purposes it is convenient to define a *tenth-value layer;* this is the thickness of a given absorber required to decrease the radiation intensity to one tenth of its original value.* According to equation

* A "half-value layer," defined in an analogous manner, is also used.

(2.60.3) this thickness is a specific property of the absorbing material and does not depend on the intensity of the incident radiation. One tenth-value layer will reduce the radiation intensity by a factor of 0.1; two tenth-value layers will reduce it by a factor of $(0.1)^2$, i.e., 0.01, and so on. In general, n such layers will decrease the gamma-ray intensity to $(0.1)^n$ or 10^{-n} of its initial value.

2.62. As seen earlier, more or less definite ranges can be ascribed to alpha and beta radiations of specified energy in a given medium. Because of the exponential nature of the absorption, however, no such range can be ascribed to gamma rays. A theoretical consequence of equation (2.60.2) is that an infinite thickness of material is required to absorb gamma radiation completely, i.e., to make $I_x = 0$. Nevertheless, in practice a finite, although not very definite, thickness will reduce the intensity to a point where it is relatively insignificant.

Absorption Coefficient and Gamma-ray Energy

2.63. The linear absorption coefficient varies with the energy of the gamma rays, because the modes of interaction of this radiation with matter, as described above, are all energy dependent in different ways. By measuring the intensity

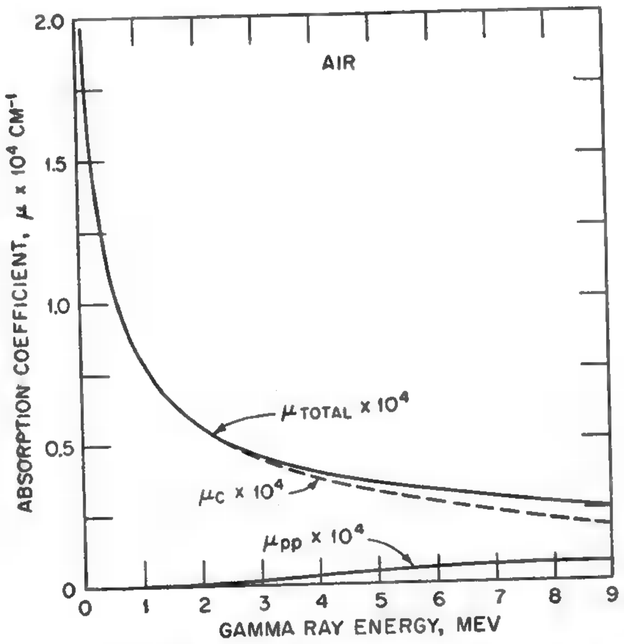


Fig. 2.10. Linear absorption coefficients of gamma rays in air

of a collimated beam of monoenergetic gamma rays before (I_0) and after (I_x) passage through a known thickness (x cm) of absorber, and using equation (2.60.3), the linear absorption coefficient (μ cm^{-1}) can be determined experimentally. By combining experimental data with theoretical considerations, it is possible to divide the observed absorption coefficient for a given gamma-ray

energy into three parts, representing the contributions made by the photoelectric effect (μ_{pe}), by the Compton effect (μ_c), and by pair production (μ_{pp}), respectively. The results for an absorbing material of low (average) atomic number (air) and for one of high atomic number (lead) are given in Figs. 2.10 and 2.11, for gamma-ray energies up to 9 Mev.

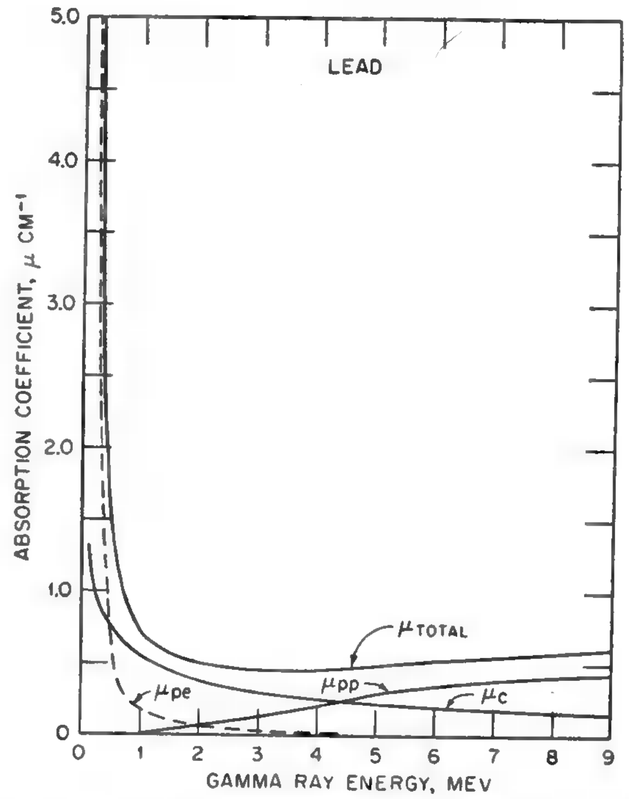


Fig. 2.11. Linear absorption coefficients of gamma rays in lead

2.64. It will be observed, in the first place, that the total absorption coefficient for lead is of the order of 10^4 times that for air; this is due to the fact that all three types of absorption interaction increase with increasing atomic number of the absorber. For air, the photoelectric effect is so small at appreciable gamma-ray energies that it makes no significant contribution; there is, consequently, no curve for μ_{pe} in Fig. 2.10, and the Compton effect is dominant up to energies of 2 Mev or more. Because both μ_{pe} and μ_c decrease with increasing energy, whereas μ_{pp} increases for energies exceeding 1.02 Mev, the total absorption coefficient may be expected to exhibit a minimum at a particular energy. Such a flat minimum is evident in Fig. 2.11 for lead, at a gamma-ray energy of about 3 to 4 Mev. It undoubtedly also exists for air as absorber, at a much higher energy.

2.65. The linear absorption coefficients of water, aluminum, iron, and lead, for gamma rays with energies from 0.5 to 5.0 Mev, are given in Table 2.4. In

TABLE 2.4. LINEAR ABSORPTION COEFFICIENTS IN cm^{-1} UNITS

Energy (Mev)	Water	Aluminum	Iron	Lead
0.5	0.090	0.23	0.63	1.7
1.0	0.067	0.16	0.44	0.77
1.5	0.057	0.14	0.40	0.57
2.0	0.048	0.12	0.33	0.51
3.0	0.038	0.090	0.30	0.47
4.0	0.033	0.082	0.27	0.48
5.0	0.030	0.074	0.24	0.48

each case the absorption coefficients are seen to decrease with increasing gamma-ray energy, at least up to energies of about 5 Mev. This means that, within this range, the higher the energy the greater the thickness of a given material required to absorb a specified fraction of the radiation.

Mass Absorption Coefficient and Thickness Density

2.66. If the ordinary (or linear) absorption coefficient, given in cm^{-1}, is divided by the density of the absorber, in grams per cm^3, the result, i.e., μ/ρ, is called the *mass absorption coefficient*, expressed in cm^2 per gram. Some values for the four absorbers referred to above are recorded in Table 2.5. It will be

TABLE 2.5. MASS ABSORPTION COEFFICIENTS IN cm^2 PER GRAM UNITS

Energy (Mev)	Water	Aluminum	Iron	Lead
0.5	0.090	0.085	0.087	0.15
1.0	0.067	0.059	0.056	0.068
1.5	0.057	0.052	0.051	0.050
2.0	0.048	0.044	0.042	0.045
3.0	0.038	0.033	0.038	0.041
4.0	0.033	0.030	0.034	0.042
5.0	0.030	0.027	0.030	0.042

observed that the mass absorption coefficients vary with the gamma-ray energy, but at any particular energy, especially in the range from 1 to 3 Mev, the values for different absorbing materials are approximately the same. This fact can be used to estimate absorption coefficients roughly when direct experimental data are lacking.

2.67. Since water has a density of unity (in gram per cm^3 units), its linear and mass absorption coefficients are identical, and so either may be taken as equal

to the (approximately) constant mass absorption coefficients of other substances. The linear absorption coefficient (μ) for any absorber, for gamma rays of a specified energy, may thus be obtained upon multiplying the absorption coefficient in water by the density of the absorber.

Example Estimate the linear absorption coefficient of concrete, density 2.8 g/cm^3, for gamma rays of 3-Mev energy.

For 3-Mev gamma rays, the (linear or mass) absorption coefficient of water is 0.038, and so the mass absorption coefficient of concrete may be taken to be 0.038 cm^2/g. By definition, this is equal to μ/ρ, and since ρ is given as 2.8 g/cm^3, it follows that

$$\mu = (0.038)(2.8)$$
$$= 0.11\ cm^{-1}.$$

2.68. Upon dividing and multiplying the exponent in equation (2.60.2) by ρ, the density of the absorber, the result is

$$I_x = I_0 e^{-(\mu/\rho)(x\rho)}, \tag{2.68.1}$$

where the first factor in the exponent is now the mass absorption coefficient defined above and the second is the thickness density of the absorber, expressed in grams per cm^2 (or mg per cm^2), as in § 2.37. Hence, when the circumstances are such that the mass absorption coefficient is (approximately) constant for different materials, the thickness density which attenuates the radiation to a specified extent will also be roughly independent of the nature of the absorber. In other words, the thickness of material required to absorb the same fraction of gamma radiation, of given energy, is approximately inversely proportional to the density of the material.

2.69. Although, as stated in § 2.62, it is not possible to specify a range for gamma rays, as it is with alpha or beta particles, a rough comparison can be made of the penetrating power of the various radiations. Since μ for 1-Mev gamma rays in water is 0.067 cm^{-1}, it is readily found from equation (2.60.2) that 70 cm (28 in.) of water would be required to decrease the radiation intensity to 1 per cent of its initial value. The range of 1-Mev beta particles in water is 0.30 cm (0.16 in.), and for 1-Mev alpha particles it is about 0.0004 cm (less than 0.2 mil).

2.70. A further discussion of absorption coefficients and the absorption of gamma radiation, in general, will be given later in connection with the design and construction of shields for the protection of personnel from the effects of nuclear radiations (Chapter X).

INTERACTION OF NEUTRONS WITH MATTER

The Production of Neutrons

2.71. Before proceeding with a description of some of the reactions between neutrons and atomic nuclei, brief reference will be made to the methods used for

producing neutrons. Not only are neutron sources of various kinds required for experimental purposes, but they also play an important role in the operation of certain reactors.

2.72. Neutrons are readily obtained by the action of alpha particles on some light elements, e.g., beryllium, boron, or lithium. A convenient source of neutrons is a mixture of an alpha emitter, such as radium or polonium, with beryllium. The reaction may be represented by

$$_4Be^9 + {}_2He^4 \rightarrow {}_6C^{12} + {}_0n^1,$$

where, as stated in Chapter I, the subscripts give the atomic number, or positive charge, and the superscripts are the respective mass numbers. The reaction may be written in the abbreviated form $Be^9(\alpha, n)C^{12}$, indicating that a Be^9 nucleus, called the *target nucleus*, interacts with an incident alpha particle (α); a neutron (n) is ejected and a C^{12} nucleus, referred to as the *recoil nucleus*, remains.

2.73. The neutrons produced by the interaction of alpha particles with beryllium have fairly high energies, ranging from 5 to 12 Mev or more, depending on the energy of the incident alpha particles. Such a source is polyenergetic, since the neutrons do not all have the same (or nearly the same) energies.

2.74. The action of gamma rays of moderate energy (about 2 Mev) on certain nuclei, notably deuterium, i.e., heavy hydrogen, and beryllium, yields essentially monoenergetic neutrons.* The reactions, which are of special interest in connection with the operation of nuclear reactors, are

$$_4Be^9 + {}_0\gamma^0 \rightarrow {}_4Be^8 \text{ (or } 2\ {}_2He^4) + {}_0n^1$$

and

$$_1H^2 + {}_0\gamma^0 \rightarrow {}_1H^1 + {}_0n^1.$$

These are described as (γ, n) reactions, since a gamma-ray photon is the incident particle and a neutron is expelled. Sources based on (γ, n) reactions are called *photoneutron sources.*

2.75. In principle, all nuclei should be capable of exhibiting the (γ, n) process, although the reaction probability, i.e., the cross section, will generally be very small. However, the energy of the gamma rays must be at least equal to the binding energy of the neutron in the given nucleus. With the exception of deuterium and beryllium, this energy is high, i.e., roughly 6 to 8 Mev, and so in practice these are the only substances used in photoneutron sources.

2.76. The minimum (or threshold) energy of the gamma rays necessary to bring about these reactions is 1.6 Mev for beryllium and 2.2 Mev for deuterium. Any energy of the gamma-ray photon in excess of the threshold value appears mainly as kinetic energy of the emitted neutron, some being carried off by the recoil nucleus. Since gamma rays from a given isotope usually have a definite energy, so also will the neutrons produced. Consequently neutrons obtained as

* This is one of the types of gamma-ray interaction, mentioned in § 2.48.

a result of a (γ, n) reaction are essentially monoenergetic, the actual energy depending on the gamma-ray energy. Possible gamma-emitting radioisotopes for use in photoneutron sources are the naturally occurring elements radium and mesothorium, and the artificially produced isotopes sodium-24, gallium-72, antimony-124, and lanthanum-140.

2.77. A simple and convenient source of approximately monoenergetic neutrons consists of a rod of antimony, containing the radioactive isotope-124, surrounded by a beryllium metal cup; it is available from the Isotopes Division of the A.E.C. When newly prepared the system emits neutrons of a roughly uniform energy of 0.03 Mev, at the rate of about 8×10^6 per sec. The antimony-124 has a half life of 60 days, and so the neutron production falls off accordingly. However, when the activity has decayed to an appreciable extent, the source can be regenerated by exposure of the antimony rod to neutrons in a nuclear reactor, thus providing a new supply of antimony-124.

Neutron Reactions: Absorption

2.78. Neutrons undergo two main types of reactions with atomic nuclei: these are (1) *absorption* (or capture), in which a neutron enters the nucleus but other particles leave,* and (2) *scattering*, in which the neutron interacts with, and transfers some or all of its energy to, the nucleus, but the neutron remains free after the process. As seen to some extent in Chapter I, both kinds of interaction play a part in the operation and control of nuclear reactors.

2.79. In considering absorption reactions it is convenient to distinguish between reactions of slow neutrons and of fast neutrons. There are four main kinds of slow-neutron reactions; these involve capture of the neutron by the target followed by either (1) the emission of gamma radiation (n, γ); (2) the ejection of an alpha particle (n, α); (3) the ejection of a proton (n, p); or (4) fission (n, f). Of these, the radiative capture, i.e., (n, γ), process is the most common, for it occurs with a wide variety of elements. The (n, α) and (n, p) reactions with slow neutrons are limited to a few isotopes of low mass number, whereas fission by slow neutrons is restricted to certain nuclei of high atomic number.

2.80. There are reasons for believing that all absorption reactions take place in two distinct stages: the first is the capture of the neutron by the target nucleus to form what is called a *compound nucleus*, as mentioned in § 1.57. This may be a nucleus of a familiar species or it may be unstable, but in any case the compound nucleus, immediately after its formation, is in a high-energy (or excited) state. Within a very short time, the excited compound nucleus undergoes the second stage of the reaction, involving either expulsion of a particle, e.g., an alpha particle or a proton, or of a gamma-ray photon, or breaking up into two more or less equal parts, i.e., fission.

* When a nucleus, e.g., uranium-235, can take part in fission as well as radiative capture, the term "neutron absorption" is used to include both reactions, whereas "neutron capture" refers to the latter only. In other cases, the terms are equivalent.

Radiative Capture Reactions

2.81. In radiative capture or (n, γ) reactions, the excited compound nucleus emits its excess energy in the form of one or more gamma-ray photons, eventually leaving the compound nucleus in its normal or ground state. The gamma rays emitted are referred to as *capture gamma rays.* As seen in § 1.58, the residual nucleus or product is isotopic with the target nucleus, but has a mass number one unit larger. Frequently this product is radioactive, exhibiting negative-beta decay, often accompanied by gamma rays. When this is the case, the occurrence of the neutron capture reaction can be detected experimentally by the resulting radioactivity. The procedure is used in various measurements with slow neutrons.

2.82. As already stated, radiative capture reactions with slow neutrons are very common; some examples, such as those with hydrogen, uranium-238, and thorium-232, have been given in Chapter I. The first of these, namely,

$$_1H^1 + {}_0n^1 \rightarrow ({}_1H^2)^* \rightarrow {}_1H^2 + \gamma,$$

is of importance, both in connection with neutron shielding and with the effects of neutrons on living tissue which consists largely of hydrogen. It will be observed that this reaction is just the reverse of that involved in the production of neutrons by the action of gamma rays on deuterium. The minimum gamma-ray energy required for the latter reaction is 2.2 Mev (§ 2.76), and the capture gamma rays produced in the (n, γ) reaction with hydrogen have the same energy. As will be seen below, this is numerically equal to the energy required to detach a neutron from a deuterium nucleus, i.e., the neutron binding energy in deuterium.

2.83. Among other nuclei which exhibit radiative capture reactions of interest, mention may be made of cadmium-113, present to the extent of 12.3 per cent in the naturally occurring element; the reaction is

$$_{48}Cd^{113} + {}_0n^1 \rightarrow ({}_{48}Cd^{114})^* \rightarrow {}_{48}Cd^{114} + \gamma.$$

The product is cadmium-114, which is a stable (nonradioactive) isotope. The cross section for this reaction is very high, and so cadmium is used as a neutron absorber for reactor control (Chapter VI). The fact that a nonradioactive product is formed is an advantage, since there is no danger in approaching a cadmium control rod after removal from the reactor.

2.84. In the reaction of slow neutrons with indium-115, the main stable isotope of this element, i.e.,

$$_{49}In^{115} + {}_0n^1 \rightarrow ({}_{49}In^{116})^* \rightarrow {}_{49}In^{116} + \gamma,$$

the product is a radioactive, negative-beta emitter with a half life of 54 min. Because of the large cross section for this radiative capture reaction and the activity of the product, it has found frequent use for the detection and measurement of neutron densities.

2.85. It may be mentioned that most of the processes whereby radioisotopes are made for various experimental or research uses involve radiative capture. A stable form of the particular (target) element is inserted in a thermal-neutron reactor, such as the Oak Ridge (X-10) or the Brookhaven reactor, for a period of time. Some of the neutrons produced by fission and slowed down by the moderator are captured. The capture is immediately followed by gamma-ray emission and the formation of an isotope of the target element. This is a type of parasitic capture (§ 1.91) for which allowance must be made in designing a reactor that is to be used, at least partly, for isotope production.

Capture Gamma Rays and Neutron Binding Energy

2.86. Some indication of the energy of the capture gamma rays may be obtained in the following manner. The formation of the compound nucleus Y from the target nucleus X by the capture of a neutron may be represented by

$$X^A + n^1 \rightarrow [Y^{A+1}]^*,$$

where the asterisk indicates an excited state of the Y nucleus. In radiative capture, the excited compound nucleus emits the excess energy and thereby passes into its normal or ground state; thus,

$$[Y^{A+1}]^* \rightarrow Y^{A+1} + E_\gamma,$$

where E_γ is the energy which appears as one or more photons of capture gamma radiation.

2.87. Next, imagine a neutron of zero kinetic energy to be removed from the ground state of the Y^{A+1} nucleus, leaving the ground state of the nucleus X^A. The energy which must be supplied is the *binding energy* (E_b) of the neutron in the compound nucleus; thus,

$$Y^{A+1} \rightarrow X^A + n^1 - E_b, \quad (2.87.1)$$

the negative sign before E_b indicating that this energy is supplied. Upon combining the three processes indicated above, it is seen that $E_\gamma = E_b$. Hence, if the excitation energy of the compound nucleus is emitted as a single gamma-ray photon, its energy will be numerically equal to E_b (Fig. 2.12*a*). However, if the energy is emitted in two or more stages (Fig. 2.12*b*), as is generally the case, the total energy $E_\gamma (= E_b)$ will be divided among the two or more photons. It should be mentioned that in the foregoing treatment the kinetic energy of the neutron was neglected. Actually E_γ will be numerically equal to E_b plus the kinetic

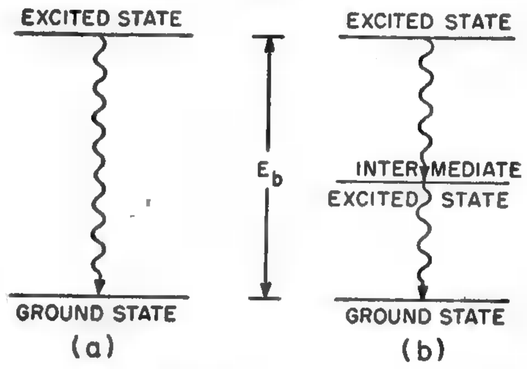


FIG. 2.12. Emission of gamma radiation by compound nucleus

energy of the incident neutron, but for *slow* neutrons the latter is so small, e.g., less than 1 ev, in comparison with E_b, e.g., 6 to 8 Mev, that it may be ignored.

2.88. The magnitude of the binding energy, and hence of E_γ, may be obtained from equation (2.87.1) by writing it in the form

$$\text{Mass of Y} = \text{Mass of X} + \text{Mass of neutron} - \text{Mass equivalent of } E_b,$$

so that

$$\text{Mass equivalent of } E_b = \text{Mass of X} + \text{Mass of neutron} - \text{Mass of Y}.$$

If the masses of the target nucleus X, of the compound nucleus Y, and of the neutron are known, the mass equivalent of E_b can be determined. The value of E_b in Mev can then be derived by means of equation (1.76.2), i.e., by multiplying the mass equivalent in atomic mass units (amu) by 931.

2.89. In some cases the masses of X and Y are available from various measurements; in other instances they can be estimated with a fair degree of accuracy by means of the following semiempirical equation, based on the liquid-drop model of the nucleus (§ 2.151). The nuclear mass M (in amu) of any species having an atomic number Z and mass number A is given by*

$$M \text{ (amu)} \times 10^3 = 1015A + 14.0A^{\frac{2}{3}} + 41.9\frac{(Z - Z_A)^2}{Z_A} - 41.9Z_A + \delta, \quad (2.89.1)$$

where

$$Z_A = \frac{66.7A}{A^{\frac{2}{3}} + 132}$$

and

$$\delta = \begin{cases} -\dfrac{35}{A^{\frac{3}{4}}} \text{ for } A \text{ even and } Z \text{ even} \\ 0 \text{ for } A \text{ odd} \\ +\dfrac{35}{A^{\frac{3}{4}}} \text{ for } A \text{ even and } Z \text{ odd.} \end{cases}$$

2.90. When using this equation to determine E_b it should be noted that the X and Y nuclei have the same value of Z and their mass numbers differ by unity. Taking the mass of the neutron as 1.00897 (§ 1.43), the mass equivalent of E_b can be calculated. Upon multiplying this result by 931, the required binding energy of the neutron is obtained in Mev units. For nearly all nuclei, other than the very lightest, E_b is found to be about 6 to 8 Mev. This is consequently the order of magnitude of the total energy of the capture gamma rays, which is frequently carried by two or more photons.

2.91. Because of the importance of the actual photon energies in connection

* For a compilation of calculated atomic masses (including masses of the orbital electrons) see N. Metropolis, "Table of Atomic Masses," Institute for Nuclear Studies, Chicago, 1948.

with certain aspects of reactor shielding and heat removal, a number of experimental studies have been made of the energy distribution (or spectrum) of capture gamma radiations. In a few cases only are these spectra known with any degree of accuracy; the results quoted in Table 2.6 are for some elements

TABLE 2.6. CAPTURE GAMMA-RAY ENERGIES OF REACTOR MATERIALS

Element	*Photon Energy* (Mev)	*Energy Per Capture* (Mev)
Aluminum	7.7	2.7
	6.0	1.4
	3.0	3.8
Beryllium	6.8	6.8
Cadmium	0 to 1	0.6
	1 to 3	0.4
	3 to 5	3.0
	5 to 7	1.0
Carbon	1 to 3	0.6
	5.0	5.0
Hydrogen	2.2	2.2
Lead	6.7	0.5
	7.4	6.8
Sodium	0 to 1	0.5
	1 to 3	1.0
	3 to 5	2.4
	5 to 7	1.8
Uranium	3.0	6.0
	1.0	0.8
Zirconium	3 to 5	4.5
	5 to 7	2.0

that are of special interest in the reactor field.* It will be observed that, for the substances listed, only with hydrogen and beryllium is the capture gamma radiation emitted as a single photon per neutron capture.

Emission of Alpha Particles or Protons

2.92. Slow-neutron reactions accompanied by the emission of a charged particle, e.g., an alpha particle (n, α) or a proton (n, p), are rare. The reason is that a positively charged particle can be expelled from a nucleus only if it has sufficient energy to overcome an electrostatic (or coulombic) potential, in addition to the energy required to detach it from the nucleus. It is only for a few

* See also, P. S. Mittelman and R. A. Liedtke, *Nucleonics*, 13, No. 5, 50 (1955).

elements of low atomic number, for which the nuclear electrostatic repulsion is small, that charged-particle emission is possible after capture of a slow neutron.

2.93. The interactions of slow neutrons with lithium-6, the less common, naturally occurring isotope of lithium, and with boron-10, the rarer, stable isotope of boron, lead to the ejection of an alpha particle. Both of these reactions have a special interest in the present connection. The (n, α) reaction with boron-10 may be written as

$$_5B^{10} + {}_0n^1 \rightarrow (_5B^{11})^* \rightarrow {}_3Li^7 + {}_2He^4,$$

where $_2He^4$ represents an alpha particle, i.e., a helium nucleus, mass number 4 and atomic number 2. The charged particles produced in this reaction are ejected in opposite directions with relatively high energy, so that they produce considerable ionization in their passage through a gas. This is the basis of a method for detecting and counting slow neutrons (Chapter V). Because boron has a high cross section for the (n, α) reaction with slow neutrons, this element, like cadmium, is used for reactor control.

2.94. The other (n, α) process which occurs readily with slow neutrons is that with lithium-6, i.e.,

$$_3Li^6 + {}_0n^1 \rightarrow (_3Li^7)^* \rightarrow {}_1H^3 + {}_2He^4.$$

The residual (recoil) nucleus is here H^3, a negative beta-active, hydrogen isotope of mass number 3, called *tritium*. This isotope has attracted attention because of its possible use in the so-called "hydrogen (fusion) bomb," and for other reasons. It can be produced by the action of slow neutrons on lithium in a nuclear reactor.

2.95. A few isotopes of low atomic number, notably helium-3, nitrogen-14, sulfur-32, and chlorine-35, undergo (n, p) reactions with slow neutrons. Of these two only need be considered. One is the reaction with helium-3, namely,

$$_2He^3 + {}_0n^1 \rightarrow (_2He^4)^* \rightarrow {}_1H^3 + {}_1H^1,$$

which, like the (n, α) reaction with boron-10, can be used for the detection of slow neutrons. The other is the (n, p) reaction with nitrogen-14; it is

$$_7N^{14} + {}_0n^1 \rightarrow (_7N^{15})^* \rightarrow {}_6C^{14} + {}_1H^1,$$

and its significance here lies in the fact that it is one way in which slow neutrons react with elements present in living tissue. The reaction product, carbon-14, is radioactive, emitting a negative beta particle, and regenerating nitrogen-14; thus,

$$_6C^{14} \rightarrow {}_{-1}\beta^0 + {}_7N^{14}.$$

Reactions with Fast Neutrons

2.96. Reactions of fast neutrons with atomic nuclei, other than scattering and fission, are not of major importance for the study of nuclear reactors. Although many such fast-neutron reactions are known, the cross sections are usually so small that they have little effect on reactor operation. One process of this type, however, should be mentioned: it is the (n, p) reaction of fast neutrons with oxygen-16, namely,

$$_8O^{16} + {}_0n^1 \rightarrow (_8O^{17})^* \rightarrow {}_1H^1 + {}_7N^{16}.$$

The product is nitrogen-16, which is beta active, having a half life of 7.3 sec, and emitting, in addition, a gamma ray of high energy (about 6 Mev). This process occurs in reactors using either air or water as coolant, since both of these materials contain oxygen. Even though most of the fissions would be produced by thermal neutrons, a considerable number of fast neutrons, liberated in fission, are present. These will interact with oxygen-16 nuclei to yield the highly radioactive, although short-lived, nitrogen-16. The decay product of the latter is ordinary oxygen-16.

2.97. Provided the energy is available, the expulsion of a charged particle from the excited compound nucleus, formed as a result of neutron capture, is more probable than the emission of radiation. Thus (n, α) and (n, p) reactions of nuclei with fast neutrons, having energies of 1 Mev or more, frequently occur more readily than the (n, γ) reaction. If neutrons of sufficiently high energy are used, two or more neutrons or protons may be expelled from the compound nucleus. For incident neutrons of about 10 Mev, such reactions as $(n, 2n)$ and (n, np) have been observed, and for still higher energies, $(n, 3n)$, $(n, 2np)$, etc., processes are possible.

Inelastic Scattering

2.98. When a fast neutron undergoes *inelastic scattering* (§ 1.51), it is first captured by the target nucleus to form a compound nucleus; a neutron of lower kinetic energy is then emitted, leaving the target nucleus in an excited state (Fig. 2.13). In other words, in an inelastic scattering collision, some (or all) of the kinetic energy of the neutron is converted into excitation (potential) energy of the target nucleus. This energy is subsequently emitted in the form of one or more photons of gamma radiation, called *inelastic-scattering gamma rays*.

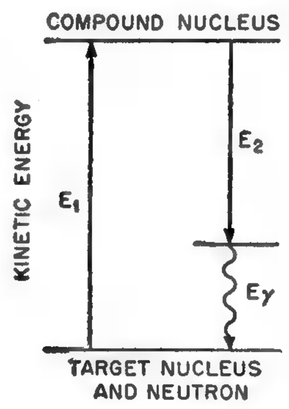


Fig. 2.13. Energy change in inelastic scattering collision

2.99. Let E_1 be the total kinetic energy of the neutron and target nucleus before collision and E_2 the kinetic energy after collision; then if E_γ is the energy emitted as gamma radiation, it follows that

$$E_1 = E_2 + E_\gamma.$$

It is evident that in inelastic scattering kinetic energy is not conserved. Nevertheless, there is conservation of momentum, so that, if E_γ were known, the mechanics of the process could be solved. This is not necessary, however, for the present purpose. What is

important to note is that E_1 must be at least equal to E_γ if inelastic scattering is to occur. Since the kinetic energy of the target nucleus is, in general, negligible in comparison with that of the neutron, it follows that, in an inelastic collision, the initial energy of the neutron must exceed the minimum excitation energy of the target nucleus.

2.100. For elements of moderate and high mass number, the minimum excitation energy, i.e., the energy of the lowest excited state above the ground state, is usually from 0.1 to 1 Mev. Hence only neutrons with energy exceeding this amount can be inelastically scattered. With decreasing mass number of the nucleus there is a general tendency for the excitation energy to increase, so that the neutrons must have higher energies if they are to undergo inelastic scattering. The threshold energy for such scattering in oxygen, for example, is about 6 Mev, and in hydrogen the process does not occur at all. Exceptions to the foregoing generalizations are the very stable, so-called, "magic number" nuclei, containing 2, 8, 20, 52, 82, and 126 neutrons or protons. Heavy nuclei of this type, e.g., lead (82 protons) and bismuth (126 neutrons), behave like light nuclei with respect to inelastic scattering.

2.101. Another general rule relating to inelastic scattering is that the relative probability of its occurrence, as against radiative capture or other processes following neutron absorption, increases with increasing neutron energy. This is because the separation (or spacing) of the excited levels of a nucleus is smaller at high excitation energies; there are consequently more excited states, in a given energy range, which the nucleus can occupy after emission of a neutron. The probability of the emission of a neutron by the compound nucleus increases correspondingly.

2.102. The energy of the inelastic-scattering gamma rays depends, of course, upon the value of E_γ in the particular case, and upon whether it is emitted as one or more photons. For inelastic scattering by elements of low mass number, the total gamma-ray energy must be high, e.g., several Mev, whereas for heavy elements it will usually be lower.

Elastic Scattering

2.103. It will be apparent from the foregoing discussion that inelastic scattering is restricted to neutrons with energies in excess of at least 0.1 Mev for collisions with the heaviest nuclei and to those with even larger energies when lighter nuclei are involved. Consequently, neutrons with energies less than about 0.1 Mev cannot lose energy as a result of inelastic collisions. The situation in regard to elastic scattering is quite different; kinetic energy is conserved, and there are no restrictions upon the transfer of this energy between the neutron and the nucleus. Provided the kinetic energy of the neutron exceeds that of the nucleus, there is a possibility that some kinetic energy will be transferred from the former to the latter, and vice versa.

2.104. Although elastic collisions may involve compound nucleus formation, as a first step, followed by emission of the neutron, the process may nevertheless be regarded essentially as a "billiard ball" type of collision, since the struck nucleus remains in its lowest energy (ground) state. The process can then be treated by the laws of classical mechanics, based on the principles of the conservation of both energy and momentum. In each collision with an essentially stationary nucleus, a neutron will transfer part of its kinetic energy to the nucleus. The amount of energy transferred will depend on the mass of the scattering nucleus and the angle through which the neutron is scattered. For a given scattering angle, the fraction of the neutron energy transferred will be greater the smaller the mass of the scattering nucleus.*

Maxwell-Boltzmann Distribution

2.105. After a sufficient number of elastic scattering collisions, the velocity of a neutron is reduced to such an extent that it has approximately the same average kinetic energy as the atoms (or molecules) of the scattering medium. The energy depends on the temperature of the medium, and so it is called *thermal energy*. Thermal neutrons are thus neutrons which are in thermal equilibrium with the atoms (or molecules) of the medium in which they are present. A particular thermal neutron undergoing collisions with the nuclei of the medium may gain or lose energy in any one collision. But, if a large number of thermal neutrons diffusing in a nonabsorbing medium are considered, there is no net energy change for all the neutrons.

2.106. In a weakly absorbing medium, the kinetic energies of thermal neutrons will be distributed statistically according to the Maxwell-Boltzmann distribution law,† as derived from the kinetic theory of gases; this may be written in the form

$$\frac{dn}{n} = \frac{2\pi}{(\pi kT)^{\frac{3}{2}}} e^{-E/kT} E^{\frac{1}{2}}\, dE, \tag{2.106.1}$$

where dn is the number of neutrons with energies in the range from E to $E + dE$, n is the total number of neutrons in the system, k is the Boltzmann constant, and T is the absolute temperature. The equation may be written in a slightly different form by letting $n(E)$ represent the number of neutrons of energy E *per unit energy interval*. Then $n(E)\, dE$ is the number of neutrons having energies in the range from E to $E + dE$, which is equivalent to dn in equation (2.106.1). The latter may thus be written as

$$\frac{n(E)}{n} = \frac{2\pi}{(\pi kT)^{\frac{3}{2}}} e^{-E/kT} E^{\frac{1}{2}}, \tag{2.106.2}$$

* The subject of elastic scattering is treated more fully in Chapter III.

† In a thermal reactor, where fast fission neutrons are being produced while slow neutrons are readily absorbed, there are positive deviations from the Maxwell-Boltzmann distribution at high energies and negative deviations at low energies (see W. J. Sturm, *Phys. Rev.*, **71**, 707 (1947)).

where the left side represents the fraction of the neutrons having energies in the range from E to $E + dE$ per unit energy interval. The right side of equation (2.106.2) can be evaluated for various E's at a given temperature, and the Maxwell-Boltzmann curve obtained in this manner, indicating the variation of $n(E)/n$ with the kinetic energy E of the neutrons, is shown in Fig. 2.14.

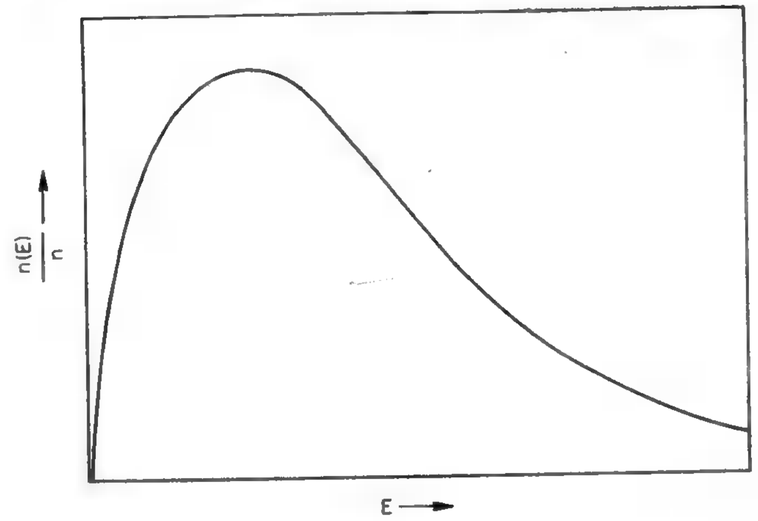


FIG. 2.14. Maxwell-Boltzmann distribution of energy

2.107. It is seen from Fig. 2.14 that, while some thermal neutrons have very small and others very large energies, a considerable proportion have energies in a fairly narrow range. It has become the practice to give the average or mean energy of thermal neutrons as kT for the particular absolute temperature T. This is actually the kinetic energy corresponding to the most probable velocity per unit velocity interval, based on the Maxwell-Boltzmann distribution. Since the Boltzmann constant k is 1.38×10^{-16} erg/°C, i.e., 8.6×10^{-5} ev/°C or 4.8×10^{-5} ev/°F, it follows that the so-called average energy of thermal neutrons is $8.6 \times 10^{-5} T_K$ or $4.8 \times 10^{-5} T_R$ ev, where T_K and T_R are the absolute temperatures of the scattering medium on the Kelvin and Rankine scales, respectively. These are the expressions which were given in § 1.53.

2.108. Since the neutron energy is kinetic, it is equal to $\frac{1}{2}mv^2$, where m is the neutron mass and v its velocity. The value of m is 1.67×10^{-24} gram, and so the neutron energy is $0.83 \times 10^{-24} v^2$ ergs or $5.2 \times 10^{-13} v^2$ ev, with v in cm per sec; consequently, it is found that

$$v = 1.4 \times 10^6 \sqrt{E} \text{ cm/sec},$$

where E is the neutron energy expressed in electron volts. If this result is combined with the expressions for the thermal-neutron energy derived above, the average speed of thermal neutrons is given as $1.3 \times 10^4 \sqrt{T_K}$ or $0.97 \times 10^4 \sqrt{T_R}$ cm/sec, as stated in § 1.54.

CROSS SECTIONS FOR NEUTRON REACTIONS

Significance of Cross Sections

2.109. The description of the interaction of neutrons with atomic nuclei can be made quantitative by means of the concept of *cross sections*, defined in general terms in § 1.63. If a given material is exposed to the action of neutrons, the rate at which any particular nuclear reaction occurs depends upon the number of neutrons, their velocity, and the number and nature of the nuclei in the specified material. The cross section of a target nucleus for any given reaction is a property of the nucleus and of the energy of the incident neutron.

2.110. Suppose a uniform, parallel beam of I neutrons/cm² impinges perpendicularly, for a given time, on a thin layer, dx cm in thickness, of a target material containing N atoms (or nuclei)/cm³, so that $N\,dx$ is the number of target nuclei per cm². Let C be the number of individual processes, e.g., neutron captures, occurring per cm². The nuclear cross section σ for a specified reaction is then defined as the average number of individual processes occurring per target nucleus per incident neutron in the beam; thus,

$$\sigma = \frac{C}{(N\,dx)I} \text{ cm}^2/\text{nucleus}. \qquad (2.110.1)$$

Because nuclear cross sections are frequently in the range of 10^{-22} to 10^{-26} cm²/nucleus, it is the general practice to express them in terms of a unit of 10^{-24} cm²/nucleus, called a *barn*. Thus, a cross section of 2.7×10^{-25} (or 0.27×10^{-24}) cm²/nucleus would be written as 0.27 barn.

2.111. The significance of the cross section may be seen by rearranging equation (2.110.1) into the form

$$(N\,dx)\sigma = \frac{C}{I}. \qquad (2.111.1)$$

If every neutron falling on the target reacted, then I would be equal to the number of nuclei taking part in the reaction; hence, the right side of equation (2.111.1) represents the fraction of the incident neutrons which succeed in reacting with the target nuclei. Thus $(N\,dx)\sigma$ may be regarded as the fraction of the surface capable of undergoing the given reaction; in other words, of 1 cm² of target surface $(N\,dx)\sigma$ cm² is effective. Since 1 cm² of the surface contains $N\,dx$ nuclei, the quantity σ cm² is the *effective area per single nucleus* for the given reaction. It is this interpretation of σ that leads to the use of the term "cross section."

Macroscopic Cross Section

2.112. The cross section σ for a particular process, which applies to a single nucleus, is frequently called the *microscopic cross section*. Since the target material contains N nuclei/cm³, the quantity $N\sigma$ is equivalent to the total cross

section of the nuclei per cm³; this is called the *macroscopic cross section* of the material for the process. Representing the latter by Σ, it is therefore defined as

$$\Sigma = N\sigma \text{ cm}^{-1}, \tag{2.112.1}$$

with dimensions of a reciprocal length.

2.113. If the target material is an element, of atomic weight A and density ρ grams/cm³, then ρ/A is the number of gram atoms per cm³. The number of atomic nuclei per cm³ is obtained upon multiplying by N_a, the Avogadro number (6.02×10^{23}), which gives the number of individual atoms (or nuclei) per gram atom; thus,

$$N = \frac{\rho}{A} N_a, \tag{2.113.1}$$

so that, from equation (2.112.1),

$$\Sigma = \frac{\rho N_a}{A} \sigma. \tag{2.113.2}$$

2.114. For a compound of molecular weight M and density ρ, the number N_i of atoms of the ith kind per cm³ is given by a modification of equation (2.113.1), namely,

$$N_i = \frac{\rho N_a}{M} \nu_i, \tag{2.114.1}$$

where ν_i is the number of atoms of the kind i in a molecule of the compound. The macroscopic cross section for this element in the given target material is then

$$\Sigma_i = N_i\sigma_i = \frac{\rho N_a}{M} \nu_i\sigma_i, \tag{2.114.2}$$

where σ_i is the corresponding microscopic cross section. For the compound, the macroscopic cross section is expressed by

$$\begin{aligned}\Sigma &= N_1\sigma_1 + N_2\sigma_2 + \cdots + N_i\sigma_i + \cdots \\ &= \frac{\rho N_a}{M} (\nu_1\sigma_1 + \nu_2\sigma_2 + \cdots \nu_i\sigma_i + \cdots).\end{aligned} \tag{2.114.3}$$

Example The microscopic cross sections for the capture of thermal neutrons by hydrogen is 0.33 barn and for oxygen 0.0002 barn. What is the macroscopic capture cross section of water for thermal neutrons?

The molecular weight (M) of water is 18 and the density (ρ) is 1 g/cm³; the molecule contains 2 atoms of hydrogen and one of oxygen. Hence, by equation (2.114.3),

$$\begin{aligned}\Sigma &= \frac{1 \times N_a}{18} (2\sigma_H + \sigma_O) \\ &= \frac{(1)(6.02 \times 10^{23})}{18} [(2)(0.33) + 0.0002](10^{-24}) \\ &= 0.022 \text{ cm}^{-1},\end{aligned}$$

recalling that the Avogadro number (N_a) is 6.02×10^{23} and the barn is 10^{-24} cm².

2.115. For a mixture, either of elements or compounds or both, which contains several different nuclear species, the macroscopic cross section is given by

$$\Sigma = N_1\sigma_1 + N_2\sigma_2 + \cdots + N_i\sigma_i + \cdots, \tag{2.115.1}$$

where the values of N_1, N_2, etc., are dependent upon the composition of the mixture as well as on the atomic (or molecular) weights and densities of the constituents.

Example In the Oak Ridge (X-10) reactor, cylindrical fuel rods (diameter 2.80 cm) of natural uranium metal (density 19.0 g/cm³) are arranged on 20-cm centers in a square lattice of graphite (density 1.62 g/cm³). The absorption cross sections for these materials are 7.42 barns and 0.0045 barn, respectively. If the reactor material may be regarded as a uniform mixture, what is its macroscopic capture cross section?

The ratio of the volume of the uranium metal to that of the whole reactor is $(\frac{1}{4}\pi)(2.80)^2$ to $(20)^2$, i.e.,

$$\frac{\text{Volume of uranium}}{\text{Total volume}} = \frac{(\frac{1}{4} \times 3.14)(2.80)^2}{400} = \frac{0.0154}{1},$$

so that 1 cm³ of the reactor contains 0.0154 cm³ of uranium and 0.9846 cm³ of carbon. Consequently there are present

$$\frac{(0.0154)(19.0)(6.02 \times 10^{23})}{238} = 7.40 \times 10^{20} \text{ uranium nuclei/cm}^3$$

and

$$\frac{(0.9846)(1.62)(6.02 \times 10^{23})}{12} = 8.00 \times 10^{22} \text{ carbon nuclei/cm}^3,$$

the atomic weight of natural uranium being taken as 238 and that of graphite (carbon) as 12.0. Then, by equation (2.115.1),

$$\begin{aligned}\Sigma &= (7.40 \times 10^{20})(7.42 \times 10^{-24}) + (8.00 \times 10^{22})(0.0045 \times 10^{-24}) \\ &= 5.85 \times 10^{-3} \text{ cm}^{-1}.\end{aligned}$$

(Because the heterogeneous reactor does not behave like a uniform system, the actual value of Σ is somewhat less than that calculated here.)

Attenuation of Neutrons: Determination of Cross Sections

2.116. In order to determine cross sections experimentally, the attenuation of a neutron beam is measured for a target material of finite thickness. Suppose a collimated beam of neutrons strikes perpendicularly a specified area of material of appreciable thickness (Fig. 2.15). Consider a thin layer, of thickness dx, parallel to the surface; then from the arguments presented above, it follows that $N\sigma\,dx$ is the fraction of the neutrons falling on this layer which react. This may be set equal to $-dI/I$, where $-dI$ is the decrease in the neutrons per cm² as the result of passing through the thickness dx of target material. Consequently,

$$-\frac{dI}{I} = N\sigma\,dx,$$

and integration over the thickness x of the material gives

$$I_x = I_0 e^{-N\sigma x} = I_0 e^{-\Sigma x}, \tag{2.116.1}$$

where I_0 is the number of incident neutrons falling on a particular area, and I_x is the number which succeed in passing through x cm of the material over the same area.*

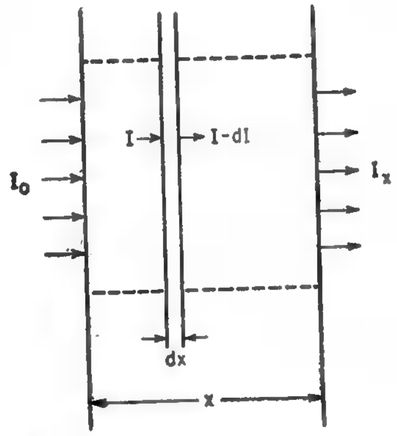


FIG. 2.15. Neutron attenuation in passage through slab

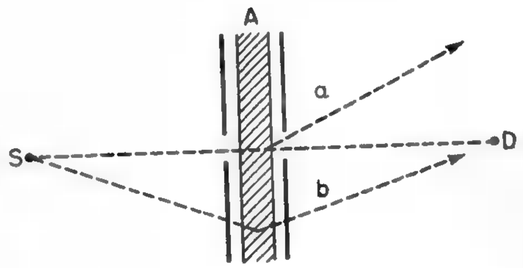


FIG. 2.16. Determination of cross sections by transmission method

2.117. The experimental arrangement for the measurement of cross sections by the transmission method consists of a neutron source S and a detector D, between which is placed a slab A of the material being investigated (Fig. 2.16). By means of a suitable collimating shield, the neutron beam passing through to the detector is restricted to a relatively small solid angle. The purpose of the shield is to prevent, as far as possible, neutrons which have been scattered in the material from reaching the detector. This will be apparent from a consideration of the dotted lines showing possible paths of scattered neutrons. If the aperture in the shield is small, the neutrons will not, in general, be scattered into the detector, as shown at a. However, if the aperture had been larger, there is a possibility that scattered neutrons would reach the detector, as indicated at b. It is assumed that the slab is not too thick, so that the neutrons are not scattered more than once within the slab; otherwise, neutrons such as a might be scattered into the detector, in spite of the small aperture in the shield.

2.118. With the arrangement suggested in Fig. 2.16, the neutrons reaching D will be those that have escaped absorption and have not been scattered. If I_0 is the neutron intensity measured at D in the absence of the slab, and I_x is the value when the slab of thickness x is interposed, then the macroscopic total cross section for absorption of all kinds and scattering can be obtained from equation (2.116.1). If N is known for the material, then the equivalent microscopic cross section can be determined.

* Attention should be called to the similarity between equation (2.116.1) and equation (2.60.2) for the absorption of gamma radiation. The linear absorption coefficient (μ) is seen to be equivalent to a macroscopic cross section; in fact, μ/N is often referred to as the (microscopic) cross section for gamma-ray absorption.

2.119. The scattering cross section can be measured by having the neutron detector in such a position that it can be reached by scattered neutrons only. This may be achieved by placing the detector at an angle approaching 90° from the incident beam; then only those neutrons which have been scattered through this angle will be counted. From the result obtained, the total number of neutrons scattered through all angles by a known thickness of material can be calculated, and hence the scattering cross section can be evaluated.

2.120. The difference between the total scattering and absorption cross section, obtained above, and the scattering cross section gives the absorption cross section for all processes. For neutrons of sufficiently high energy, where neutron absorption is of the same order as, or smaller than, scattering, this method is inapplicable, since it involves the difference of two almost equal numbers containing experimental inaccuracies. Another procedure for determining absorption cross sections is available when the nuclear species being studied forms a radioactive product as a result of interaction with neutrons. The cross section for the particular capture process can then be calculated from the measured activity (see § 2.203).

RATES OF NEUTRON REACTIONS

2.121. Consider a neutron beam in which n is the *neutron density*, i.e., the number of neutrons per cm³; if v is the neutron velocity, then nv is the number of neutrons falling on 1 cm² of target material per sec. Since σ cm² is the effective area per single nucleus, for a given reaction or reactions (§ 2.111), then Σ is the effective area (in cm²) of all the nuclei per cm³ of target. Hence, the product Σnv gives the number of interactions (between neutrons and nuclei) per cm³ of target material per sec. Since each nuclear interaction involves one neutron, it follows that

$$\text{Rate of neutron interaction} = \Sigma nv \text{ neutrons/(cm}^3\text{)(sec)}. \tag{2.121.1}$$

This is a result of considerable importance, for it gives the number of neutrons per second involved in any interaction (or interactions) with 1 cm³ of material for which Σ is the macroscopic cross section. It is sometimes written in a slightly different form by introducing the *neutron flux*, in place of the neutron density. The neutron flux, represented by ϕ, is equal to the product nv, expressed as neutrons per cm² per sec, so that equation (2.121.1) becomes

$$\text{Rate of neutron interaction} = \Sigma\phi \text{ neutrons/(cm}^3\text{)(sec)}, \tag{2.121.2}$$

an expression which will find frequent use in later sections.

Cross Section and Mean Free Path

2.122. An alternative approach to the determination of neutron reaction rates is to consider the *mean free path*, λ, i.e., the average total (or scalar) distance a neutron will travel before undergoing a particular interaction. Since the velocity v is the distance a neutron travels per second, the average number of interactions per neutron per second is v/λ. For a beam containing n neutrons/cm^3, the number of interactions per cm^3 per second is thus nv/λ. In other words,

$$\text{Rate of neutron interaction} = \frac{nv}{\lambda} \text{ neutrons/(cm}^3\text{)(sec)}. \qquad (2.122.1)$$

Upon comparing equations (2.121.1) and (2.122.1), it is seen that

$$\lambda = \frac{1}{\Sigma}, \qquad (2.122.2)$$

so that the neutron mean free path for a specified reaction is the reciprocal of the macroscopic cross section for that reaction. If Σ is expressed in cm^{-1}, as it usually is, then λ will be in cm.

Example Using the result of the preceding example, determine the average distance traveled by a thermal neutron in the Oak Ridge (X-10) reactor before it is absorbed.

According to the preceding example, Σ in this reactor is 5.85×10^{-3} cm^{-1}; hence,

$$\lambda = \frac{1}{5.85 \times 10^{-3}} = 171 \text{ cm}.$$

A thermal neutron consequently travels, on the average, a total distance of 171 cm (67 in.) before it is captured by either uranium or graphite. (Because of the heterogeneous arrangement the actual mean free path in the reactor is larger.)

2.123. Equation (2.116.1) for the attenuation of neutrons by passage through a target material can be rewritten in an alternative form upon replacing Σ by $1/\lambda$, in accordance with equation (2.122.2); the result is

$$I_x = I_0 e^{-x/\lambda}. \qquad (2.123.1)$$

If the thickness x of the material is equal to λ, then $I_x/I_0 = 1/e$. Hence, after traversing a thickness λ of the target material, a fraction $1/e$ of the incident neutrons have not been involved in the particular process under consideration. If λ is the mean free path for absorption, then the intensity of the neutron beam is reduced to a fraction $1/e$ of its initial value after passage through this thickness of absorber.*

2.124. When a neutron can take part in several different processes with a given target nucleus, e.g., (n, γ), (n, α), (n, f), inelastic scattering, and elastic scattering, there will be a specific cross section and mean free path for each process. The equations derived above are quite general and will apply to any

* In this connection the distance λ is sometimes referred to as the "relaxation length" for nuclear radiations (see § 10.64).

reaction in which neutrons are involved. It is possible to define a total absorption cross section, or even a total cross section for both absorption and scattering, which is the sum of the individual cross sections. An equation of the form of (2.116.1) will be applicable, and I_x/I_0 will give the fraction of the initial neutrons escaping interaction.

Polyenergetic Neutron Systems

2.125. In the derivations given above, it has been assumed, for simplicity, that all neutrons have the same velocity. Since cross sections, in particular those for neutron absorption, are strongly energy dependent, there will be a different value for each neutron energy. For a polyenergetic beam of neutrons, equation (2.116.1) would thus become more complicated. If it were possible to divide the initial neutron beam into a number of definite energy groups, indicated by the subscripts 1, 2, 3, etc., then the initial intensity I_0 would be given by

$$I_0 = I_{01} + I_{02} + \cdots + I_{0i} + \cdots,$$

and then, for the ith group,

$$I_{xi} = I_{0i} e^{-N\sigma_i x},$$

so that

$$I_x = \sum_i I_{xi} = \sum_i I_{0i} e^{-N\sigma_i x}, \qquad (2.125.1)$$

where the large sigma implies summation over all values of i.

2.126. In deriving this result it has been tacitly assumed, as it was, in fact, in equation (2.116.1), that, when passing through the thickness x of medium, the neutron velocities (or energies) do not change appreciably, so that the cross sections remain constant. This condition is particularly applicable to thermal neutrons, and, since there is then a fairly definite (Maxwell-Boltzmann) energy distribution, it is possible to derive an average cross section, as will be shown below. In this case, equation (2.125.1) can be considerably simplified.

2.127. If, in a polyenergetic neutron system, $n(E)$ is the density of neutrons of energy E per unit energy interval, then, as seen in § 2.106, $n(E)\,dE$ is the number in the energy range from E to $E + dE$. The total neutron flux ϕ, for neutrons of all energies (or velocities), is then given by

$$\phi = \int_0^\infty n(E)v\,dE \text{ neutrons/(cm}^2\text{)(sec)}, \qquad (2.127.1)$$

where the integration limits of zero and infinity are formal only and are meant to imply that integration is carried out over the whole range of neutron energies. The velocity v is that corresponding to the kinetic energy E. The rate of interaction of the polyenergetic neutron system can then be expressed, using equation (2.121.2), as

$$\text{Rate of neutron interaction} = \int_0^\infty \Sigma(E)n(E)v\,dE \text{ neutrons/(cm}^3\text{)(sec)}, \qquad (2.127.2)$$

where $\Sigma(E)$ is the macroscopic cross section for the process for neutrons of energy E.

2.128. For the given system, it is possible to define an average macroscopic cross section $\bar{\Sigma}$ so that

$$\text{Rate of neutron interaction} = \bar{\Sigma}\phi \text{ neutrons/(cm}^3\text{)(sec)}, \qquad (2.128.1)$$

where ϕ is the total neutron flux derived above. Upon combining equations (2.127.1), (2.127.2), and (2.128.1), it is seen that

$$\bar{\Sigma} = \frac{\int_0^\infty \Sigma(E)n(E)v\,dE}{\int_0^\infty n(E)v\,dE} \text{ cm}^{-1}. \qquad (2.128.2)$$

For thermal neutrons having a Maxwell-Boltzmann distribution, $n(E)$ in the foregoing equations would be defined by equation (2.106.2), with n equal to the total number of neutrons per cm^3.

2.129. In order to be able to perform the indicated integrations, however, it is necessary to express $\Sigma(E)$ as a function of the neutron energy E. For many absorbers the $1/v$ law (§ 1.65) is obeyed in the thermal-neutron region; that is, $\Sigma(E)$ is a linear function of $1/v$ and, consequently, of $1/E^{\frac{1}{2}}$. Upon combining this result with equations (2.106.2) and (2.128.2), it is found that $\bar{\Sigma}_{th}$ for the absorption of thermal neutrons is equal to the cross section for neutrons of energy $4kT/\pi$, i.e., with energy $4/\pi$ times the so-called average thermal neutron energy kT (§ 2.107). Alternatively, if Σ_{kT} is the absorption cross section for neutrons having actual energy kT, it follows from the $1/E^{\frac{1}{2}}$ law that

$$\bar{\Sigma}_{th} = \frac{\sqrt{\pi}}{2}\Sigma_{kT} \quad \text{or} \quad \bar{\Sigma}_{th} = 0.886\Sigma_{kT}. \qquad (2.129.1)$$

This average value for the absorption cross section of thermal neutrons may be used in equation (2.128.1) to give the rate of interaction of these neutrons, or in equation (2.116.1) to determine their attenuation in passing through a thickness x of absorber.

VARIATION OF CROSS SECTIONS WITH NEUTRON ENERGY

Experimental Results

2.130. The problem of the complete determination of cross sections for neutron reactions is a very complex one; not only do the values depend on the neutron energy, but they vary from one isotope to another of the same element and change with the nature of the reaction. In many instances, one reaction predominates, and this simplifies the situation; the same is true when one particular isotope has a much larger cross section than others of the same element. Most measurements have been made with naturally occurring materials, and the data are useful for calculations of the rates of processes occurring in nuclear reactors. For some elements, however, cross sections have also been derived for individual isotopes, such as uranium-235, uranium-238, etc.

2.131. For many elements, especially those of mass number exceeding 100, an examination of the variation of the absorption cross sections with neutron energy reveals the existence of three regions.* There is, first, a low-energy region, where the cross section decreases steadily with increasing neutron energy. The absorption cross section σ_a then varies inversely as the square root of the neutron energy and, since the energy is kinetic in nature, σ_a is inversely proportional to the neutron velocity. This is called the $1/v$ *region*, and the neutrons are said to obey the $1/v$ law.

2.132. Following the $1/v$ region for slow neutrons, the elements under consideration exhibit a *resonance region*, usually for neutrons of roughly 0.1 to 1000 ev

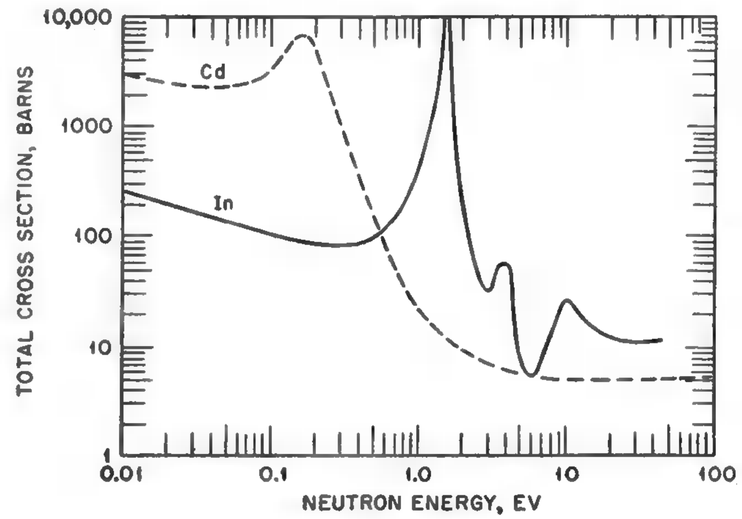


Fig. 2.17. Cross sections of cadmium and indium as function of neutron energy

energy. This region is characterized by the occurrence of peaks where the absorption cross section rises fairly sharply to high values for certain neutron energies and then falls again. Some elements, e.g., cadmium and rhodium, have only one resonance peak, while others, such as indium, silver, gold, and uranium-238, have two or more peaks. The cross sections of cadmium and indium, as functions of the neutron energy, are shown in Fig. 2.17 and those for natural uranium in Fig. 2.18. The scales are logarithmic in both directions, and so the $1/v$ region, at the left of the figure, appears as a straight line. The resonance peak for cadmium occurs at 0.18 ev and the absorption cross section is then about 7200 barns for the natural element.† The main peak for indium is for

* Since scattering cross sections are usually small, the total cross section, i.e., absorption and scattering, shows the same trend.

† The resonance is due to the cadmium-113 isotope present to the extent of 12.3 per cent; for this isotope in pure form the cross section is about 20,000 barns.

neutrons of 1.44 ev energy, and the cross section exceeds 20,000 barns. There are also lower resonance peaks at about 4 and 10 ev. The highest peak for uranium is at 7 ev and the cross section is about 4000 barns.

2.133. Since marked resonance peaks are found in regions of relatively low neutron energies and with elements of high mass number, the reactions taking place are usually of the radiative capture (n, γ) type. The only exceptions are the few nuclear species which also undergo fission following absorption of slow neutrons. As pointed out earlier, the heavier elements would require neutrons having energies in the Mev range for other processes. The behavior of certain substances of low mass number undergoing (n, α) reactions with slow neutrons will be referred to in § 2.138.

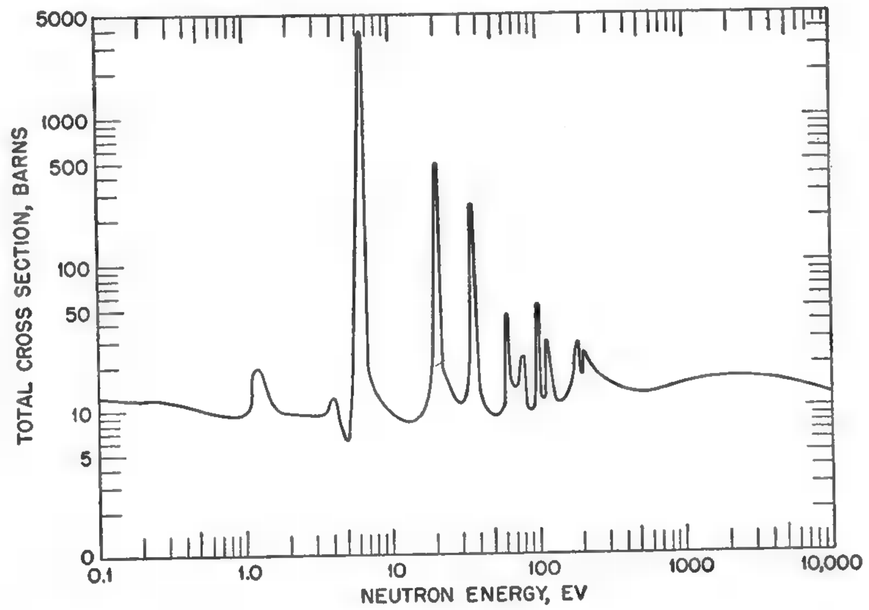


Fig. 2.18. Cross section of natural uranium as function of neutron energy

2.134. Beyond the resonance region, the nuclear cross sections decrease steadily with increasing neutron energy. This represents what is called the *fast-neutron region*. The cross sections are usually low, being less than 10 barns in most cases and becoming even smaller for energies of the order of 0.1 Mev or more. The absorption cross sections are then similar in magnitude to the geometrical cross section of the nucleus, i.e., 2 to 3 barns (§ 2.147). Some nuclei exhibit resonance behavior in the fast-neutron region, but the peaks are not very high and are of little immediate significance.

Theoretical Interpretation: Resonance Absorption

2.135. There are good reasons for believing that an atomic nucleus can be stable (or quasistable) only when its energy corresponds to that of a particular

quantum state. Every nucleus has several such states, the lowest being the stable or ground state, whereas the others are various excited quantum states or energy levels.* When a nucleus captures a neutron and the energy of the resulting compound nucleus is equal (or very close) to one of the quantum states of this nucleus, the probability of capture is exceptionally high. This is the accepted explanation of resonance absorption with its associated large capture cross sections.

2.136. The resonance effect may be illustrated by reference to Fig. 2.19; the lines at the right indicate (schematically) the quantum levels of the compound nucleus. The line marked E_0, at the left, represents the energy of the compound nucleus formed when the target nucleus absorbs a neutron of zero kinetic energy. As seen in § 2.87, the excitation energy of the compound nucleus at E_0 is just (numerically) equal to the binding energy of the neutron. An examination of Fig. 2.19 shows that the energy E_0 does not correspond to any of the quantum levels of the compound nucleus. However, if the neutron has a certain amount of kinetic energy, sufficient to bring the energy of the compound nucleus up to E_1, the nucleus will be in one of its quantum states. Thus, when the neutron has kinetic energy $E_1 - E_0$, resonance absorption occurs, and the nuclear cross section is exceptionally large. There will, similarly, be resonance absorption for neutrons of kinetic energy $E_2 - E_0$, so that the total energy E_2 is equal to that of another quantum level of the compound nucleus, as seen in Fig. 2.19.†

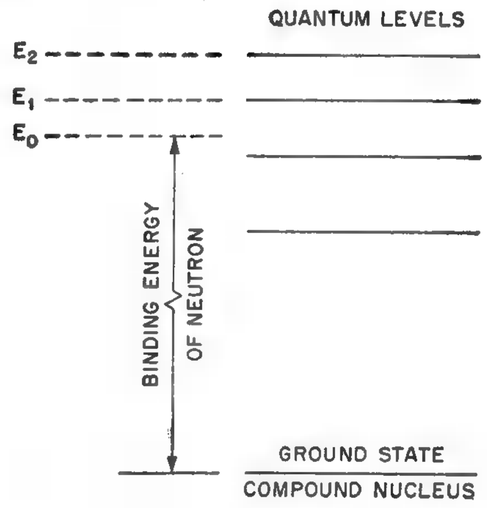


Fig. 2.19. Formation of excited compound nucleus and quantum levels

Elements of Low Mass Number

2.137. It is important to point out that not all elements show the type of behavior described above. Most elements of low mass number, as well as several of high mass number, do not exhibit resonance absorption in the region from 0.11 to 1000 ev, at least not to any appreciable extent. There may be reso-

* For small excitation energies, as are of interest here, the quantum levels are fairly widely spaced and distinct; at high energies the spacing becomes very much closer and the quantum states are virtually continuous.

† A detailed treatment of resonance absorption is given by G. Breit and E. P. Wigner, *Phys. Rev.*, **49**, 519 (1936), who have derived an expression for the cross section as a function of neutron energy applicable, particularly, in the region of a resonance peak; see also S. Glasstone and M. C. Edlund, "The Elements of Nuclear Reactor Theory," D. Van Nostrand Co. Inc., 1952, pp. 27–30.

nances at high neutron energies, but these are usually not very significant. The total neutron cross sections, including both absorption and scattering, are small, being of the order of a few barns over the whole energy range, from thermal values to several Mev.

2.138. There are, however, outstanding examples of exceptional behavior among light elements that deserve special mention. These are lithium-6 and boron-10, both of which undergo the (n, α) reaction with slow neutrons. The dependence on neutron energy of the cross section for the (n, α) reaction with ordinary boron, i.e., a mixture of boron-10 and -11, is shown in Fig. 2.20. The logarithmic plot of σ against E is essentially linear from about 0.01 ev to 0.1 Mev, although the data in the figure go only to 1000 ev. This means that the $1/v$

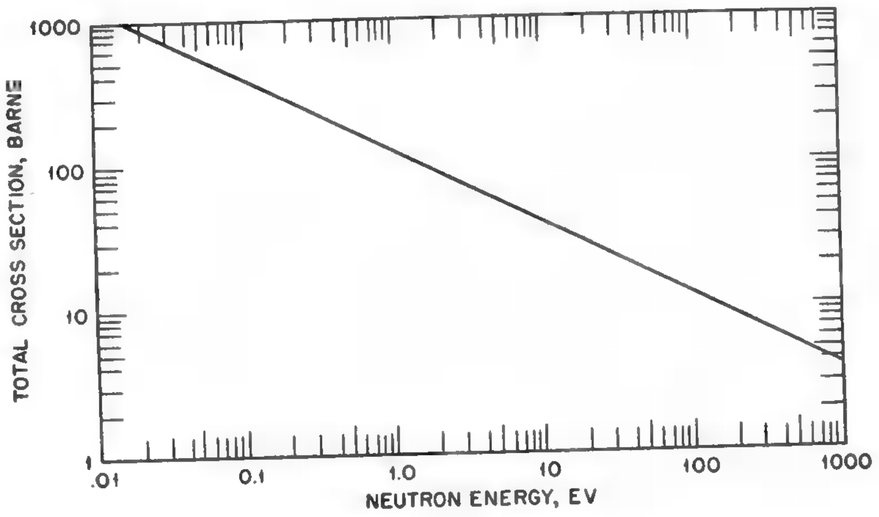


FIG. 2.20. Cross sections of boron as function of neutron energy

law holds at energies well beyond those found for (n, γ) reactions. It will be noted, too, that, while the cross sections are high for low-energy neutrons, there is, nevertheless, no indication of a resonance region.

2.139. The sharpness of a resonance peak is related theoretically (see § 2.136, footnote) to what is called the "level width" of the quantum state involved. The level width is a measure of the energy spread of the particular quantum state, and this is inversely proportional to the mean lifetime of the compound nucleus in that state. If this state is relatively stable, i.e., if the decomposition probability is fairly low and the lifetime relatively long, the level width will be small and a sharp resonance peak will be observed. This is the case with the (n, γ) reactions considered above.

2.140. However, when the compound nucleus formed by neutron capture has sufficient excitation energy to make possible the emission of a charged particle, the probability that this process will occur is large. The energy spread of the

particular quantum state is then considerable and the resonance concept, based on a more or less definite value of the energy, ceases to have any significance. In these circumstances the $1/v$ law for the variation of the cross section with neutron energy, which is characteristic of the low-energy region, will extend over a considerable range, as it does for the (n, α) reactions with lithium-6 and boron-10.

Scattering Cross Sections

2.141. With the exception of hydrogen, for which the value is as high as 20 barns in the unbound state,* and deuterium the scattering cross sections of nearly all elements lie in the range from about 2 to 10 barns for neutrons of low energy. In most cases the cross sections do not vary markedly with neutron energy, although there may be a general tendency for the values to decrease for neutrons of high energy. At very high neutron energies, the scattering cross sections are of the same magnitude as the geometrical cross sections of the respective nuclei, as will be shown below.

2.142. At low neutron energies, elastic scattering is believed to be largely due to what is called *resonance scattering*. The target nucleus captures a neutron to form a compound nucleus; the latter then expels the neutron, leaving the target nucleus in its ground state but having exchanged some kinetic energy with the neutron. It is to be expected from theoretical considerations that the resonance scattering cross section should be independent of neutron energy when the latter is less than the resonance value. At resonance a moderate maximum should occur and subsequently the cross section should decrease steadily with increasing neutron energy.

2.143. In addition to resonance elastic scattering, another type, called *potential scattering*, has been postulated from theoretical considerations, although for practical purposes the two types are taken together. Potential scattering does not necessarily involve compound nucleus formation, but, in the language of wave mechanics, it results from interaction of the neutron wave with the potential at the nuclear surface. With increasing neutron energy (or velocity) the potential scattering cross section decreases steadily. Except, perhaps, at or near a resonance, potential scattering is generally more important than resonance scattering.

2.144. In view of the fact that inelastic scattering is observed, in general, only for neutron energies exceeding about 0.1 Mev, and then only for the heavier elements, it is evident that the cross section will be zero for low and moderate neutron energies. At a particular energy, which depends upon the nature of the nuclear species, the inelastic scattering cross section becomes appreciable and then decreases gradually with increasing neutron energy. Hydrogen is excep-

* In the bound state, as in solid paraffin, the scattering cross section increases to 80 barns for neutrons of very low energy, e.g., 0.01 ev.

tional in its behavior, for, as stated earlier, it does not, as far as is known, exhibit inelastic scattering of neutrons.

2.145. It will be apparent from the foregoing remarks that the total scattering cross section, including both elastic and inelastic scattering, will rarely exceed a few barns and will not vary much with neutron energy. The very lightest nuclear species, namely, hydrogen and deuterium, have somewhat higher scattering cross sections, and, in the bound state, the values increase with energy at very low neutron energies.

Cross Sections at High Neutron Energies

2.146. At high neutron energies, generally in excess of 1 Mev, the cross sections for both compound nucleus formation and for elastic scattering approach the geometrical cross section of the nucleus. This means that, since resonance scattering is negligible, the total cross section for absorption and inelastic scattering, on the one hand, and for elastic scattering, on the other hand, each tend toward πR^2, where R is the nuclear radius. The total cross section (σ_t) for interaction of the nucleus with a high-energy neutron thus approaches the limit of

$$\sigma_t = 2\pi R^2. \tag{2.146.1}$$

2.147. It has been found in various ways that the radii of atomic nuclei, except those of very low mass number, may be represented approximately by

$$R \approx 1.4 \times 10^{-13} A^{\frac{1}{3}} \text{ cm},$$

where A is the mass number of the nucleus. Hence the total microscopic cross section is given by

$$\begin{aligned} \sigma_t &\approx 1.25 \times 10^{-25} A^{\frac{2}{3}} \text{ cm}^2/\text{nucleus} \\ &\approx 0.125 A^{\frac{2}{3}} \text{ barns}. \end{aligned} \tag{2.147.1}$$

For an element of mass number 125, for example, the limiting value of σ_t would be about 3 barns; for a mass number of 216 it would be 4.5 barns.

Example Estimate the average distance a fast neutron will travel in uranium-235 metal (density 19.0 g/cm^3) before it undergoes a nuclear interaction.

The distance required is the mean free path, taking into consideration all possible interactions. According to equation (2.147.1), the total cross section is $(1.25 \times 10^{-25})(235^{\frac{2}{3}})$, i.e., 4.76×10^{-24} cm^2/nucleus. Then, from equation (2.113.2), the corresponding macroscopic cross section is

$$\Sigma_t = \frac{(19.0)(6.02 \times 10^{23})(4.76 \times 10^{-24})}{235} = 0.23 \text{ cm}^{-1},$$

so that the mean free path, equal to $1/\Sigma_t$, is about 4.4 cm (1.75 in.). (This calculation is the basis of the rough value of 5 cm, or 2 in., used for the migration length of fast neutrons in § 1.113.)

Thermal-neutron Cross Sections*

2.148. The scattering (σ_s) and absorption (σ_a) cross sections for thermal neutrons of a few elements, of interest in connection with nuclear reactors, are given in Table 2.7; data for other elements will be found in the Appendix. It will be

TABLE 2.7. THERMAL-NEUTRON CROSS SECTIONS

Element	σ_s (barns)	σ_a (barns)	Element	σ_s (barns)	σ_a (barns)
Aluminum	1.4	0.21	Hydrogen	20–80	0.33
Beryllium	7	0.009	Indium	2.2	190
Bismuth	9	0.032	Iron	11	2.4
Boron	4	750	Lead	11	0.17
Cadmium	7	2400	Nitrogen	10	1.78
Carbon	4.8	0.0045	Oxygen	4.2	0.0002
Deuterium	5.4	0.00046	Sodium	4.0	0.50
Fluorine	4.1	0.009	Uranium	8.2	7.42
Helium	0.8	~ 0	Zirconium	8	0.18

noted that the scattering cross section for hydrogen is quoted as 20 to 80 barns; as seen above, the value depends upon whether the atom is in the free state or bound in a molecule and also upon the precise thermal-neutron energy. For unbound hydrogen, the appropriate value is 20 barns, but for the bound state it increases to about 80 barns, as the neutron energy decreases from about 0.1 ev.

2.149. The large absorption cross sections of boron and cadmium account for the use of these elements in neutron-absorbing control rods. The small values for deuterium (heavy hydrogen), beryllium, and carbon (graphite), on the other hand, point up the advantage of these substances as moderators. Similarly, the relatively small absorption cross sections of aluminum, zirconium, and iron (stainless steel) makes possible their use as structural materials. Lead and bismuth, and magic number nuclei (§ 2.100), in general, have low absorption cross sections. Although of little value for structural purposes, these elements may find other applications in reactor design, e.g., in coolants.

THE FISSION PROCESS

Mechanism of Nuclear Fission

2.150. A number of aspects of the fission process were discussed in Chapter I, since they were necessary for a general description of the fundamental principles

* For a compilation of neutron cross section data for all energies, see The A.E.C. Neutron Cross Section Advisory Group, Report AECU-2040 (and supplements), to be issued in revised form in 1955 as Report BNL-325; also "Nuclear Data," National Bureau of Standards Circular 499 (and supplements), and New Nuclear Data sections of *Nuclear Science Abstracts*, U. S. Government Printing Office.

of nuclear reactors. Some of these subjects will be examined further here, and certain additional topics will be considered. In the fission reaction a nucleus absorbs a neutron of appropriate speed, and the resulting compound nucleus breaks up into two more or less equal parts, with the liberation of a considerable amount of energy. At the same time two or three neutrons are usually emitted, mostly, although not entirely, at the instant of fission. Instantaneous gamma radiation also accompanies the fission process, and beta particles and gamma rays are produced over a period of time as the radioactive fission products decay.

2.151. A helpful insight into some of the characteristics of the fission process is obtained by the use of the liquid-drop model of the atomic nucleus. It should be clearly understood that this is merely a model which makes possible an examination of the behavior of an atomic nucleus in understandable physical terms and is not meant to represent its actual properties. The liquid-drop model has been used in various ways, e.g., to calculate nuclear masses and binding energies [cf. equation (2.89.1)], and it will be applied here to the problem of nuclear fission.

2.152. Consider a drop of liquid to which a force is applied so that it is set into oscillation; the system passes through a series of stages, of which the most significant are shown in Fig. 2.21. The drop is at first spherical, as at A; it is

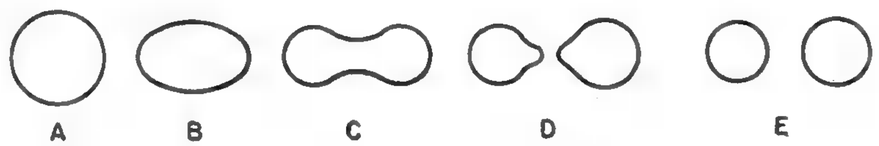


FIG. 2.21. Liquid-drop model of fission

then elongated into an ellipsoid, as at B. If insufficient energy is available to overcome the force of surface tension, the drop will return to its original form; but if the deforming force is sufficiently large, the liquid drop acquires a shape similar to a dumbbell, as at C. Once it has reached this stage it is unlikely to return to the spherical form, but it will rather split into two droplets. These will, at first, be somewhat deformed, as at D, but finally they will become spherical, as at E.

2.153. The situation in nuclear fission may be thought of as being analogous to that just considered. A target nucleus absorbs a neutron and forms an excited compound nucleus. The excitation energy of the latter is then equal to the binding energy of the neutron plus any kinetic energy the neutron may have had before its capture (§ 2.87). As a result of this excess energy, the compound nucleus may be considered to undergo a series of oscillations, in the course of which it passes through a phase similar to Fig. 2.21, B. If the oscillation energy is insufficient to cause further deformation beyond B, the intranuclear attractive forces will compel the nucleus to return to its original form. The excess energy

is then removed by the expulsion of a particle or a gamma-ray photon from the excited compound nucleus.

2.154. If, however, the compound nucleus has enough energy to permit it to pass into the dumbbell phase (Fig. 2.21, C), the restoration of the initial state A becomes improbable. Consequently, from C the system passes rapidly to D and then to E, representing fission into two separate nuclei — the fission fragments — which are propelled in opposite directions. The excess energy which the compound nucleus must have in order to permit it to deform into the state C is called the *critical energy* for fission. If this energy is available, e.g., as excitation energy following neutron capture, then fission will usually occur. If this amount of energy is not available, fission is not possible, at least not at any appreciable rate.

2.155. According to calculations based on the liquid-drop model,* the critical energy for fission decreases as the value of Z^2/A increases, Z being the atomic number and A the mass number of the nucleus. When Z^2/A is less than about 35, the critical energy is so large that neutrons of very high energy would be required to cause fission. But for nuclei having Z^2/A values of more than 35, the critical energy is down to 7 Mev or less (Table 2.8); this is the order of magnitude of the excitation energy resulting from the capture of a neutron of moderate energy. The substances in Table 2.8 are, therefore, those of immediate interest. It may be mentioned that when Z^2/A exceeds 45 or so, e.g., a nucleus of atomic number 108 and mass number 260, the critical energy for fission would be zero; such a nucleus could not exist, for, if it did, it would undergo instantaneous fission.

TABLE 2.8. CRITICAL ENERGY FOR FISSION

Target Nucleus	Z^2/A	*Critical Energy* (Mev)	*Neutron Binding Energy* (Mev)
Thorium-232	34.9	7.5	5.4
Uranium-238	35.5	7.0	5.5
Uranium-235	36.0	6.5	6.8
Uranium-233	36.4	6.0	7.0
Plutonium-239	37.0	5.0	6.6

SLOW- AND FAST-NEUTRON FISSION

2.156. The excitation energy E_{ex} of the compound nucleus is equal to the sum of the neutron binding energy E_b and the kinetic energy E of the neutron. If E_c represents the critical energy defined above, then the condition for fission is that

$$E_{ex} = E_b + E \geq E_c. \tag{2.156.1}$$

The binding energy can be obtained from the nuclear (or isotopic) masses, if they are known, or from equation (2.89.1), as described in § 2.88. The results are

* N. Bohr and J. A. Wheeler, *Phys. Rev.*, **56**, 426 (1939); S. Frankel and N. Metropolis, *ibid.*, **72**, 914 (1947).

recorded in Table 2.8. Since the critical energies have been evaluated, approximately at least, on the basis of the liquid-drop model, the minimum neutron energy E necessary to produce fission of a particular nucleus can be estimated roughly from equation (2.156.1).

2.157. For the absorption of a neutron by uranium-238, to form the compound nucleus uranium-239, it is seen from Table 2.8 that E_b is 5.5 Mev, whereas E_c is about 7.0 Mev. It follows, therefore, that the neutron would need $7.0 - 5.5 =$ 1.5 Mev of kinetic energy to make fission of uranium-238 possible. Experiments have shown that the minimum energy is about 1.1 Mev. The discrepancy between the observed (1.1 Mev) and the calculated (1.5 Mev) neutron energies is, no doubt, due partly to the inexact nature of the calculations. A somewhat similar situation holds for thorium-232 which requires neutrons with energies in excess of 1 Mev to cause fission.

2.158. On the other hand, it is evident that for uranium-233, uranium-235, and plutonium-239 the value of E_b is in each case larger than the critical energy E_c. In other words, the excitation energy of the compound nucleus, even for a neutron having no kinetic energy, is greater than the critical energy for fission. Slow neutrons should thus be capable of causing fission of uranium-233, uranium-235, and plutonium-239, as indeed they are.

2.159. The difference in behavior of uranium-235 and uranium-238, for example, with regard to fission is due mainly to the fact that the excitation energy of the compound nucleus, i.e., the neutron binding energy, is considerably larger in the former case, namely, 6.8 Mev against 5.5 Mev. A detailed examination of the binding energy calculations shows that the responsibility for the difference lies in the last term in equation (2.89.1), which can be positive, zero, or negative, according to the nuclear type. The general conclusions reached are summarized in Table 2.9, which shows whether the neutron binding energy is (relatively)

TABLE 2.9. NEUTRON BINDING ENERGIES AND NUCLEAR TYPE

Target Nucleus		*Neutron Binding Energy*	*Fissionable by Slow Neutrons*
Mass No.	*Atomic No.*		
Odd	Even	Large	Yes
Even	Even	Small	No
Odd	Odd	Small	No
Even	Odd	Large	Yes

large or small for target nuclei of various types. It is seen that the binding energy is large for target nuclei of odd mass number and even atomic number, or vice versa. Provided their Z^2/A values exceed about 35, such nuclei should be capable of undergoing fission by slow neutrons.* Isotopes of even mass number and odd atomic number are very unstable and do not exist in nature to any appreciable extent. The only species of practical interest for the utilization of

* Slow-neutron fission is possible with nuclei having even numbers of both neutrons and protons, provided Z^2/A is sufficiently large, e.g., ${}_{96}Cm^{240}$ for which Z^2/A is 38.4.

nuclear energy are those with odd mass numbers and even atomic numbers, namely, ${}_{92}U^{233}$, ${}_{92}U^{235}$, and ${}_{94}Pu^{239}$. Others do exist, but they have such short half lives that they are of no practical interest.

Fission Cross Sections

2.160. The fission cross sections for uranium-233, uranium-235, and plutonium-239, which are fissionable by slow neutrons, may be expected to vary with energy in a manner similar to that for the radiative capture processes, as already described. At low neutron energies the cross sections vary with energy in accordance with the $1/v$ law, and at high energies the values become quite small, approaching the geometrical cross sections (§ 2.146).

2.161. The thermal-neutron cross sections of uranium-235, plutonium-239, uranium-238, and natural uranium, which are important for nuclear reactor work, are recorded in Table 2.10. It is seen that 84 per cent of the thermal neu-

TABLE 2.10. THERMAL-NEUTRON CROSS SECTIONS OF FISSIONABLE NUCLEI

Nucleus	*Fission* (barns)	*Radiative Capture* (barns)	*Total Absorption* (barns)
Uranium-235	549	101	650
Plutonium-239	664	361	1225
Uranium-238	0	2.8	2.8
Uranium (natural)	3.92	3.5	7.42

trons absorbed by uranium-235 cause fission, whereas only 65 per cent of those absorbed by plutonium-239 are effective in this connection.

Fission Rate and Reactor Power

2.162. It was seen in § 2.121 that the rate of any reaction involving monoenergetic neutrons can be expressed as $\Sigma\phi$, where Σ is the macroscopic cross section for the process and ϕ is the flux of the monoenergetic neutrons. As applied to fission this becomes

$$\text{Fission rate} = \Sigma_f\phi \text{ fissions}/(\text{cm}^3)(\text{sec}), \qquad (2.162.1)$$

where

$$\Sigma_f = N\sigma_f \quad \text{and} \quad \phi = nv,$$

N being the number of fissionable nuclei per cm³ and σ_f cm²/nucleus the fission cross section; n is the neutron density, i.e., neutrons per cm³, and v is the neutron velocity in cm per sec. Although equation (2.162.1) is strictly applicable to fissions by neutrons with a single velocity, it can be used to determine the fission rate in a thermal reactor; σ_f is then the average fission cross section for thermal neutrons (§ 2.129), and v is their mean velocity. In general, if σ_f and v are appropriate average values, equation (2.162.1) can still be used to determine the fission rate in a reactor in which appreciable fissions are caused by neutrons of various energies.

2.163. In a reactor of volume V cm³, there will occur $V\Sigma_f\phi$ fissions/sec. Since it requires a fission rate of 3.1×10^{10} fissions/sec to produce 1 watt of power (§ 1.86), provided the reactor has been operating for some time, it follows that the power P of a nuclear reactor in watts is given by

$$P = \frac{V\Sigma_f\phi}{3.1 \times 10^{10}} \text{ watts.} \qquad (2.163.1)$$

Thus, for a reactor of given active volume, in which fissions are due to neutrons in a specified range, the power is proportional to the product of the macroscopic fission cross section and the neutron flux. As the reactor operates, Σ_f, which is equal to $N\sigma_f$, must decrease somewhat, since there is a gradual decrease in N, the number per cm³ of fissionable nuclei. For practical purposes, however, N and, hence, Σ_f, may be regarded as constant; consequently, the power level is proportional to the neutron flux. The power output of a nuclear reactor is thus altered by changing the neutron flux or neutron density. Similarly, the neutron flux or neutron density is widely used as a measure of the power level.

2.164. For practical use, equation (2.163.1) may be put into an alternative form as follows: the product $V \times N$ gives the total number of fissionable nuclei, and, upon division by the Avogadro number (6.02×10^{23}) and multiplication by the atomic weight (235) of the fissionable substance, the result is the mass, g, of the latter in grams. Thus,

$$\text{Mass of fissionable material} = g = \frac{235VN}{6.02 \times 10^{23}} \text{ grams.}$$

From equation (2.163.1), it follows that

$$P = \frac{VN\sigma_f\phi}{3.1 \times 10^{10}}$$

$$= 8.3 \times 10^{10} g\sigma_f\phi \text{ watts.} \qquad (2.164.1)$$

If the mass of uranium-235 is expressed as w lb, then equation (2.164.1) becomes

$$P = 3.7 \times 10^{13} w\sigma_f\phi \text{ watts.}^* \qquad (2.164.2)$$

Since the results are given to two significant figures only, they will be equally applicable when uranium-233 or plutonium-239 is the fissionable material.

Example A typical experimental thermal (uranium-graphite) reactor of the Brookhaven type contains about 50 tons of natural uranium metal. If the average thermal-neutron flux is 2×10^{12} neutrons/(cm²)(sec), what is the average power of the reactor?

Since natural uranium contains 0.7 per cent of uranium-235, the amount of the latter in the reactor is (50)(2000)(0.007) = 700 lb; this is the value of w in equation (2.164.2). The fission cross section σ_f is roughly 550×10^{-24} cm²/nucleus (Table 2.10), and the flux ϕ is 2×10^{12} neutrons/(cm²)(sec). Consequently,

$$P = (3.7 \times 10^{13})(700)(550 \times 10^{-24})(2 \times 10^{12})$$

$$= 28 \times 10^6 \text{ watts,}$$

so that the average power is about 28,000 kw. (This is the approximate operating power of the Brookhaven reactor.)

* It should be noted that in this equation σ_f is expressed in cm²/nucleus; if the value is in barns, then the corresponding equation would be $P = 3.7 \times 10^{-11} w\sigma_f\phi$ watts.

Fission Neutrons

2.165. As stated in § 1.174, the neutrons released in fission can be divided into two categories, namely, *prompt neutrons* and *delayed neutrons*. The former are released within a period of 10^{-14} sec (or less) of the instant of fission, somewhat in the following manner. The excited compound nucleus formed by the capture of a neutron first breaks up into two nuclear fragments, each of which has too many neutrons for stability, as well as the excess (excitation) energy, about 6 to 8 Mev, required for the expulsion of a neutron. The excited, unstable nuclear fragment frequently expels one or more neutrons — the prompt neutrons — within a very short time of its formation. Some of the instantaneous gamma rays accompanying fission are apparently emitted at the same time.

2.166. The energy spectrum of the prompt fission neutrons has been studied from about 0.075 to 17 Mev, and it has been found that the results can be fitted quite adequately to the formula

$$n(E) = \sqrt{\frac{2}{\pi e}} \sinh\sqrt{2E}\, e^{-E} = 0.484 \sinh\sqrt{2E}\, e^{-E}, \qquad (2.166.1)$$

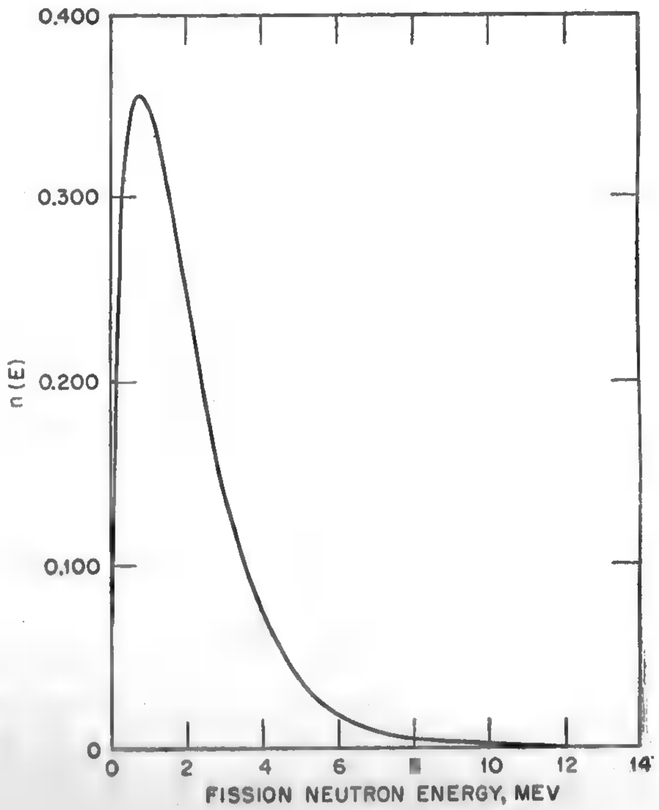


Fig. 2.22. Fission neutron energy spectrum

where $n(E)$ is the number of neutrons with energy E per unit energy interval, for each neutron emitted.* This expression applies to fission of both uranium-235 and plutonium-239. In Fig. 2.22 the values of $n(E)$ given by equation (2.166.1) are plotted as a function of E. It is seen that, although most of the prompt neutrons have energies between 1 and 2 Mev, there are nevertheless some with energies in excess of 10 Mev. This is brought out more clearly in Fig. 2.23, in which $n(E)$ is plotted on a logarithmic scale. The fact that a small proportion of fission neutrons have energies of 10 Mev or more is not important for the design of a nuclear reactor itself, but it may be significant in connection with the problems of shielding.

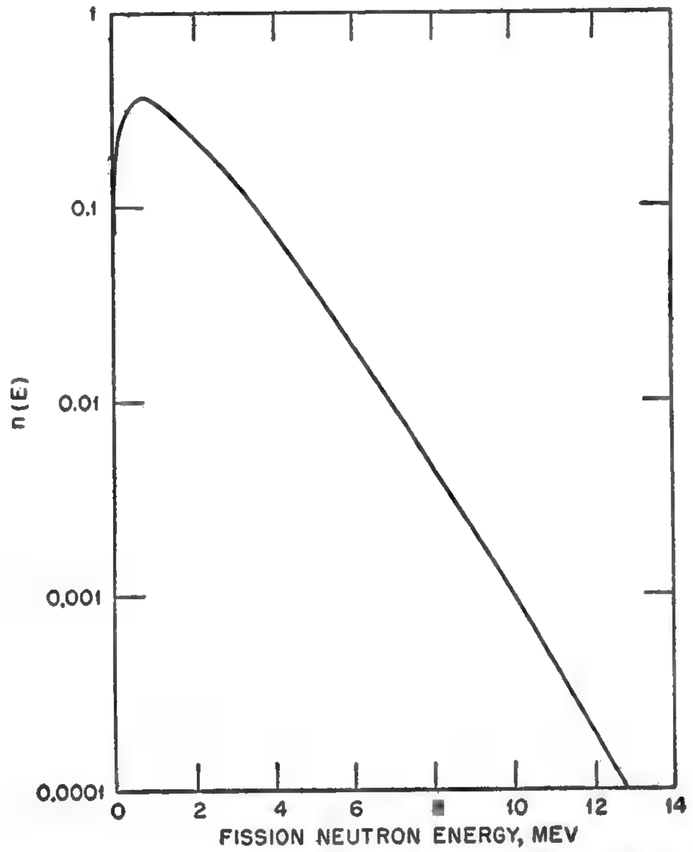


FIG. 2.23. Fission neutron energy spectrum on logarithmic scale

2.167. The delayed neutrons accompanying fission of uranium-233, uranium-235, and plutonium-239 fall into five major groups, each of which is characterized by a definite rate of decay. By observing the decay of the delayed neutrons after fission has ceased, it has been possible to associate a specific half life with each group. The results for thermal-neutron fission are given in Table 2.11,†

* B. E. Watt, Report AECD-3073; *Phys. Rev.*, **87**, 1037 (1952).

† See D. J. Hughes, *et al.*, *Phys. Rev.*, **73**, 111 (1948).

TABLE 2.11. CHARACTERISTICS OF DELAYED FISSION NEUTRONS IN THERMAL FISSION

Half Life (sec)	Fraction of Fission Neutrons Delayed			Energy (Mev)
	U-233	U-235	Pu-239	
0.43	1.8×10^{-4}	8.5×10^{-4}	4.0×10^{-4}	0.40
1.52	6.2	24.1	10.5	0.67
4.51	8.6	21.3	11.2	0.41
22.0	5.8	16.6	9.4	0.57
55.6	1.8	2.5	1.2	0.25
Total	0.0024	0.0073	0.0036	

together with the fraction which the group constitutes of the total (prompt and delayed) fission neutrons and the neutron energy, in each case. Allowing for minor groups not included in the table, the total fraction of the fission neutrons which are delayed is about 0.0025 for uranium-233, 0.0075 for uranium-235, and 0.0038 for plutonium-239. The energies of the delayed neutrons are seen to be fairly small compared with those of most of the prompt neutrons.

2.168. The fact that the delayed neutrons have definite decay half lives suggests that their origin is associated with the disintegration of certain radioisotopes. The 55.6-sec half life has been definitely associated with the decay of bromine-87 and the 22.0-sec half life with iodine-137. The following scheme has been proposed to account for the 55.6-sec delayed neutron group, and a similar mechanism is probably applicable in the other cases.

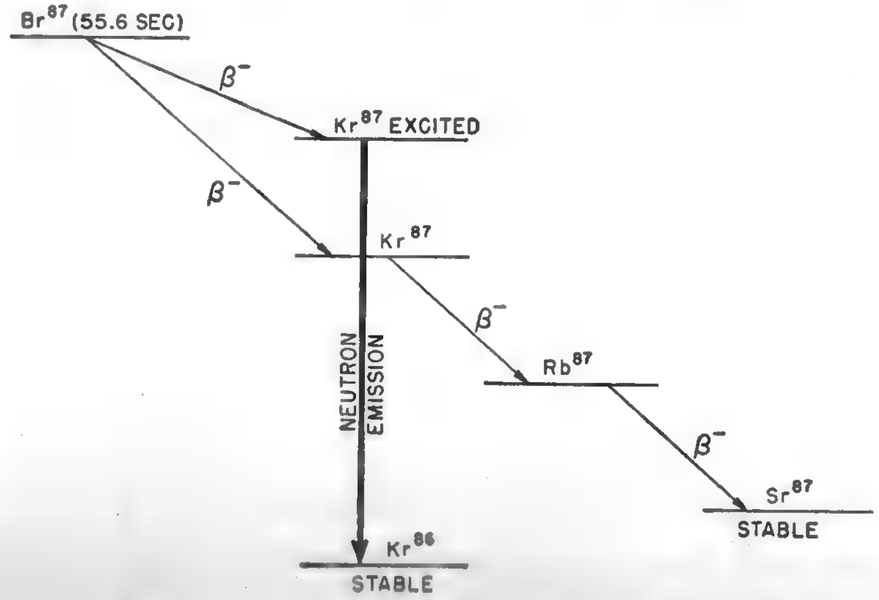


FIG. 2.24. Mechanism of delayed neutron emission

2.169. One of the products of fission is the beta emitter, bromine-87, decaying with a half life of 55.6 sec to produce krypton-87. The latter is evidently formed in a highly excited state, with sufficient energy (about 6 Mev) to permit it *immediately* to eject a neutron and leave a stable krypton-86 nucleus (Fig. 2.24). Any excess energy available, after removal of the neutron, appears as kinetic energy of the latter, as indicated in the last column of Table 2.11. The observed rate of emission of the delayed neutrons is determined by the rate of formation of the neutron emitter, krypton-87, and this is dependent on the rate of decay of the bromine-87. The latter, which is called the *precursor* of the 55.6-sec group of delayed neutrons, decays in an exponential manner, with a half life of 55.6 sec, and so the neutron emission falls off at the same rate.

2.170. The precursor of the 22.0-sec delayed neutron group is probably iodine-137; this has a half life of 22.0 sec, forming xenon-137 which instantaneously expels a neutron. Here again, the emission of the neutron is delayed because the rate of formation of the xenon-137, from which it originates, depends on the rate of decay of the precursor, iodine-137. The other three groups of delayed neutrons are probably produced in an analogous manner, although the precursors in these cases have not yet been identified.

Fission Gamma Radiation

2.171. The gamma radiation from fission is sometimes classified as prompt and delayed, as with neutron emission. Part of the prompt or instantaneous gamma radiation is produced at the same time as the prompt neutrons, the remainder being emitted by radioactive fission fragments of very short life. The delayed gamma rays, on the other hand, are those emitted over an extended period by the many longer-lived radioactive products resulting from fission. The latter will be considered more fully later.

2.172. Only two sets of measurements have been reported of the gamma radiation accompanying fission of uranium-235 (Table 2.12).* Although they agree in indicating that the total prompt gamma-ray energy is about 5 Mev, there is a considerable difference in the estimates of the average energy per photon. For various calculations it is found satisfactory to assume that the fission gamma radiation is emitted as five photons of 1-Mev energy.

TABLE 2.12. ENERGY OF FISSION GAMMA RADIATION

Investigators	*Total Energy per Fission* (Mev)	*Average Energy per Photon* (Mev)
Deutsch and Rotblat	5.1 ± 0.3	1
Kinsey, Hanna, and Van Patter	4.6 ± 0.1	2.5

* M. Deutsch and H. Rotblat, Report AECD-3179; B. B. Kinsey, G. C. Hanna, and D. M. Van Patter, *Canad. J. Res.*, **26A**, 79 (1948).

Fission Products

2.173. A detailed study of the slow-neutron fission of uranium-235 has shown that the compound nucleus splits up in more than 40 different ways, yielding over 80 primary fission products (or fission fragments).* The range of mass numbers of the products is from 72, probably an isotope of zinc (atomic number 30), to 160, possibly an isotope of gadolinium (atomic number 64). In Fig. 2.25 the mass numbers of the products of uranium-235 fission are plotted against the corresponding *fission yields*, the fission yield being defined as the proportion (or percentage) of the total nuclear fissions that form products of a given mass number. Since the observed fission yields range from 10^{-5} to over 6 per cent, they are plotted on a logarithmic scale. It should be noted that, as two nuclei result

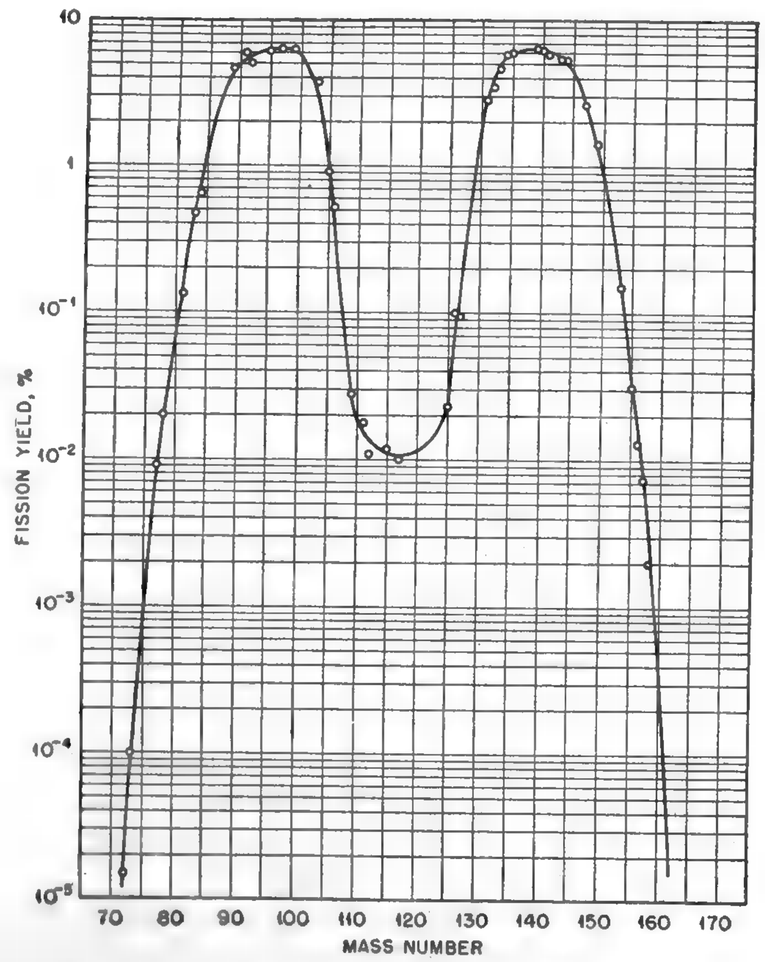


FIG. 2.25. Fission yield as function of mass number

* The terms "primary fission products" or "fission fragments" refer to the nuclei formed directly in fission. The general expression "fission products" is applied to the mixture of primary products and the various species formed by their radioactive decay.

from each act of fission, the total yield for all mass numbers adds up to 200 per cent. Incidentally, the reason why mass numbers, rather than atomic numbers, are considered is that most of the fission fragments are radioactive, decaying by the loss of a negative-beta particle. The atomic numbers, consequently, change with time, but the mass numbers are unaffected by beta decay (§ 1.58).

2.174. An examination of Fig. 2.25 shows that the masses of nearly all the fission products fall into two broad groups, a "light" group, with mass numbers from 80 to 110, and a "heavy" group, with mass numbers from 125 to 155. There are some products between and outside these ranges, but altogether they represent no more than a few per cent or so of the fissions. The most probable type of fission, comprising nearly 6.4 per cent of the total, gives products with mass numbers 95 and 139. It is apparent that the slow-neutron fission of uranium-235 is unsymmetrical in the great majority of cases. Curves similar to that in Fig. 2.25, but with maxima and minima displaced slightly from those for uranium-235, have been obtained for the slow-neutron fission of uranium-233 and plutonium-239.

2.175. Corresponding to the distribution of mass numbers among the fission products, there has been observed a distribution of kinetic energy. Conservation of momentum requires that the products of smaller mass numbers should have the higher velocities and, hence, higher energies. Consequently, two distinct kinetic-energy groups, analogous to the two mass-number groups, have been detected; the kinetic energies are approximately 61 Mev for the most abundant member of the heavy group and 93 Mev for the corresponding one in the light group. The ratio of 93 to 61 is about 1.5, and this is very close, as it should be if momentum is conserved, to the ratio of the mass numbers for maximum yield, i.e., 139 to 95, or 1.46.

2.176. During the fission process many of the orbital electrons of the atom undergoing fission are ejected, with the result that the fission fragments are highly charged. The lighter fragments carry an average positive charge of about 20 units, whereas the heavy fragments carry some 22 positive charges. Such particles, moving at speeds of the order of 10^9 cm/sec, are able to produce considerable ionization in their passage through matter. Because of their large mass and charge the specific ionization is high, and their range is, therefore, relatively short (§ 2.27). Thus, the ranges of the light and heavy groups of fission fragments have been found to be about 2.5 cm (1 in.) and 1.9 cm (0.8 in.), respectively, in air. These are roughly of the same order of magnitude as the ranges of alpha particles from radioactive sources.

2.177. The range of the fission fragments in various materials is important for reactor design, since it is necessary to prevent their escape from the fuel element into the surrounding medium, e.g., water, which might be the coolant or liquid moderator. In aluminum, for example, such as is used for cladding the uranium fuel in the Oak Ridge (X-10) and Brookhaven reactors, the range of the fission fragments has been found experimentally to be about 1.4×10^{-3} cm, i.e., about

0.55 thousandth of an inch.* As a working approximation, the range of the fission fragments in any medium may be taken to be the same as that of 4-Mev alpha particles.

Radioactivity of Fission Fragments

2.178. Nearly all of the fission fragments are radioactive, emitting negative beta particles. The immediate decay products are also usually radioactive and, although some decay chains are longer and some shorter, each fragment is followed, on the average, by three stages of decay before a stable species is formed. Since there are some 80 different radioisotopes, produced in fission and each is, on the average, the precursor of two others, there are over 200 radioactive species present among the fission products after a short time.

2.179. A large proportion of the radioactive fission products emit gamma rays, in addition to beta particles. These represent the so-called delayed fission gamma radiation. Most of it is of moderate energy, less than about 2 Mev, but a few of the fission products expel photons of higher energy. The latter are of interest for many shielding problems and for other reasons. Table 2.13 † gives a list of fission fragments and their daughters which are known to emit relatively high-energy gamma rays.‡ The fission yields refer to the fission of uranium-235 (see Fig. 2.25), and the "gamma yield" is the proportion of the

TABLE 2.13. HIGH-ENERGY GAMMA RAYS FROM FISSION PRODUCTS

Radioactive Species	Half Life*	Fission Yield (per cent)	Gamma Yield (per cent)	Energy (Mev)
Ru^{106}, Rh^{106}	1y, 30s	0.48	2	2.9
Ce^{144}, Pr^{144}	290d, 17.5m	5.3	small	>2.2
			2	2.2
			2	2.6
Eu^{156}	15.4d	0.013	60	2.0
Ba^{140}, La^{140}	12.8d, 40h	6.1	3.2	2.5
Te^{132}, I^{132}	77h, 2.4h	4.5	2.7	2.0
Te^{131m}, Te^{131}	30h, 25m	0.45	21.6	>2.2
I^{135}	6.7h	5.6	1.95	2.4
			4	1.8
Kr^{89}, Rb^{89}	2.77h, 17.8m	3.1	15	2.8
			19–34	1.9

* y = year, d = day, h = hour, m = minute, s = second.

* See E. Segrè and C. Wiegand, *Phys. Rev.*, **70**, 808 (1946).

† W. K. Ergen, unpublished; see also, J. Moteff, *Nucleonics*, **13**, No. 5, 28 (1955).

‡ Two entries in one line refer to a fission fragment and its daughter; the latter emits the high-energy gamma ray. The symbol Te^{131m} refers to an excited (high-energy) or metastable state of this isotope.

decays that are accompanied by gamma radiation in each case. The product of the percentages in the third and fourth columns gives the number of gamma photons of each particular kind per 10,000 fissions. Where the energy is given as >2.2 Mev, it means that the value is known to be greater than the photoneutron threshold energy for deuterium (§ 2.76) but has not been determined more precisely.

Decay of Fission Products: Energy Release and Radioactivity

2.180. While it is possible, theoretically, to express the rate of decay of the complex fission product mixture in terms of the fission yields and the radioactive constants of the various isotopes, it is quite impractical because of the complexity of the resulting expressions. There are, however, two problems of interest arising in reactor work which may be considered. One is to estimate the rate of energy (heat) release from the fission product mixture due to emission of beta particles and gamma rays; and the other is to determine the amount of a particular fission product present in a reactor at various times.

2.181. Although the half lives of the fission products range from a small fraction of a second up to a million years, it has been found that the rate of emission of beta particles and of gamma-ray photons can be stated by means of a simple empirical expression which is probably accurate within a factor of two or less.* From about 10 sec up to several weeks after fission has taken place, these rates (per fission) are given approximately as follows:

Rate of emission of beta radiation

$$= 3.8 \times 10^{-6} t^{-1.2} \text{ particles/(sec)(fission)}, \qquad (2.181.1)$$

Rate of emission of gamma radiation

$$= 1.9 \times 10^{-6} t^{-1.2} \text{ photons/(sec)(fission)}, \qquad (2.181.2)$$

where t is the time after fission in *days*. The mean energy of the beta particles from the fission products is about 0.4 Mev and that of the gamma-ray photons

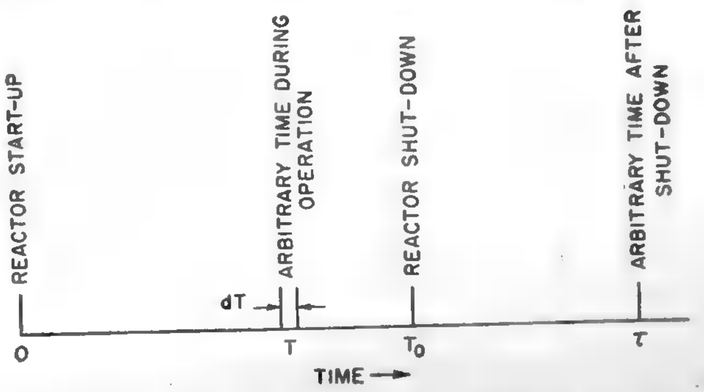


Fig. 2.26. Calculation of rate of beta and gamma energy release by fission products.

* K. Way and E. P. Wigner, *Phys. Rev.*, 73, 1318 (1948).

is about 0.7 Mev, so that

Rate of emission of beta and gamma energy

$$= 2.7 \times 10^{-6} t^{-1.2} \text{ Mev/(sec)(fission)}. \qquad (2.181.3)$$

2.182. Suppose a reactor has been operating at a constant power of P watts for a period T_0 days, and that it is required to determine the rate of emission of beta and gamma energy from the fission products after shut-down. Consider a small interval dT at a time T days after start-up (Fig. 2.26). The rate of emission of beta and gamma energy, at a time τ days after start-up, due to fissions occurring during the interval dT is then given by equation (2.181.3) as $2.7 \times 10^{-6}(\tau - T)^{-1.2}$ Mev/(sec)(fission). Since 3.1×10^{10} fissions/sec or 2.68×10^{15} fissions/day yield 1 watt of power, the number of fissions occurring in the reactor (operating at P watts) during the time interval dT days is $2.68 \times 10^{15} P\, dT$. It follows, therefore, that

Rate of emission of beta and gamma energy at time τ, due to fissions in interval dT

$$\approx (2.7 \times 10^{-6})\,(2.68 \times 10^{15}) P(\tau - T)^{-1.2}\, dT \text{ Mev/sec}$$

$$\approx 7.3 \times 10^{9} P(\tau - T)^{-1.2}\, dT \text{ Mev/sec}.$$

Consequently, for fissions occurring during the whole of the period of the reactor operation, i.e., from $T = T_0$ to $T = 0$,

Rate of emission of beta and gamma energy at time τ

$$\approx 7.3 \times 10^{9} P \int_{T_0}^{0} (\tau - T)^{-1.2}\, dT$$

$$\approx 3.7 \times 10^{10} P[(\tau - T_0)^{-0.2} - \tau^{-0.2}] \text{ Mev/sec}.$$

Upon introducing the conversion factor, 1 Mev = 1.6×10^{-13} watt-sec (§ 1.86), it is seen that

Rate of emission of beta and gamma energy at time τ

$$\approx 5.9 \times 10^{-3} P[(\tau - T_0)^{-0.2} - \tau^{-0.2}] \text{ watt}, \qquad (2.182.1)$$

where P is the power of the reactor in watts. It will be noted from Fig. 2.26 that $\tau - T_0$ is the time in days after shut-down, sometimes called the "cooling" time, and τ is the time after start-up at which this equation gives the rate of energy release due to the decay of the fission products.

2.183. The results of equation (2.182.1) have been expressed in graphical form, as in Fig. 2.27. This gives the total beta and gamma power per watt of reactor power as a function of the time $\tau - T_0$ after shut-down, for several different operating periods T_0. The actual rate of energy release is obtained upon multiplying the value given by Fig. 2.27 by P, the reactor power in watts.

2.184. If it is desired to know the activity of the fission products in curies (§ 2.14), an approximate estimate can be made from equation (2.181.1), giving the rate of emission of beta particles, utilizing the fact that every particle corre-

sponds to one nuclear disintegration. Since 1 curie is equivalent to 3.7×10^{10} dis/sec, it follows that

$$\text{Activity of fission products} \approx 1.03 \times 10^{-16} t^{-1.2} \text{ curie/fission},$$

where t is the time in days after fission. By means of arguments similar to those in the preceding paragraphs, it is found that, for a reactor which has been operating at a power of P watts for T_0 days,

$$\text{Activity of fission products at time } \tau \approx 1.4P[(\tau - T_0)^{-0.2} - \tau^{-0.2}] \text{ curies.} \quad (2.184.1)$$

It is seen that if a reactor has been operating for some time at a power of the order of megawatts, the activity of the fission products will, soon after shut-down, be of the order of megacuries.

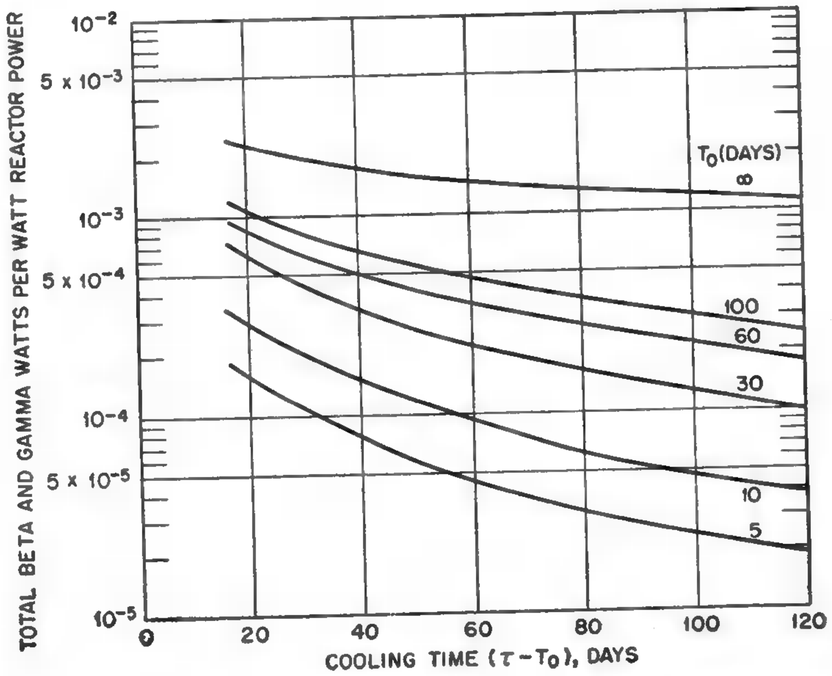


FIG. 2.27. Total beta-particle and gamma-ray power from fission products

2.185. It will be noted that equations (2.182.1) and (2.184.1), giving the rate of energy release and the radioactivity, respectively, of the fission products after a reactor is shut down, differ only in the numerical factors. Consequently, Fig. 2.27 can also be used to give the activity in curies; all that is necessary is to multiply the beta-gamma power in watts by the factor $1.4/(5.9 \times 10^{-3})$, i.e., by 240. The results are also expressed graphically in Fig. 7.5.*

* See also, K. Thornton and W. Houghton, *J. Appl. Phys.*, **24**, 1374 (1953); R. L. Koontz and A. A. Jarrett, *Nucleonics*, **12**, No. 6, 26 (1954).

ACCUMULATION OF SPECIFIC FISSION PRODUCTS

2.186. For certain purposes, e.g., the study of reactor poisoning by fission products (see Chapter IV) or the extraction of particular products, it is required to know how specific fission products accumulate during reactor operation and decay after shut-down. In the reactor a radioactive fission fragment A will be formed directly by fission and will decay to form the product B; the latter will decay in turn to yield C, and so on; thus,

$$\text{Fission} \rightarrow \text{A} \xrightarrow{\beta^-} \text{B} \xrightarrow{\beta^-} \text{C} \xrightarrow{\beta^-}.$$

In addition to undergoing radioactive decay, the substances A, B, C, etc., may capture neutrons while the reactor is operating. By taking into account the various processes, it is possible to set up differential equations for the rate of change of concentration of the specific products, A, B, C, etc., and these can be solved relatively simply in certain cases. In the following treatment it is supposed that the neutrons either are monoenergetic or are in thermal equilibrium with their environment. Alternatively, it may be assumed that proper average values for the cross sections and neutron flux are used.

2.187. Consider, first, the fission fragment A; if γ_A is its fission yield, expressed as a fraction, then the rate of formation of A due to fission is equal to $\gamma_A \Sigma_f \phi$ nuclei/(cm³)(sec), where $\Sigma_f \phi$ is the number of fissions/(cm³)(sec). If A is the number of nuclei/cm³ of A at any instant, and λ_A is its radioactive decay constant, the natural rate of radioactive decay will be $-\lambda_A A$ nuclei/(cm³)(sec). In addition, A nuclei will be removed from the system as a result of neutron capture at a rate equal to $-\sigma_A \phi A$ nuclei/(cm³)(sec), where σ_A is the capture cross section. It is thus possible to write

$$\frac{dA}{dT} = -\lambda_A A - \sigma_A \phi A + \gamma_A \Sigma_f \phi \quad (2.187.1)$$

for the net rate of increase of A nuclei with time.

2.188. When the reactor has been operating for some time, at constant power, so that $\Sigma_f \phi$ is constant, an equilibrium will be reached, when the rate of formation of A is equal to its rate of disappearance; dA/dT will then be zero. Indicating the equilibrium concentration of A as A_0, it follows from equation (2.187.1) that

$$A_0 = \frac{\gamma_A \Sigma_f \phi}{\lambda_A + \sigma_A \phi} = \frac{\gamma_A \Sigma_f \phi}{\lambda_A^*}, \quad (2.188.1)$$

where λ_A^*, defined by

$$\lambda_A^* \equiv \lambda_A + \sigma_A \phi, \quad (2.188.2)$$

may be regarded as the effective removal (decay and capture) constant of A in the reactor, provided the neutron flux remains constant. The equilibrium amount of A is seen to depend on the neutron flux in the reactor.

2.189. An expression for $A(T)$, the concentration of A nuclei at any time T, can be obtained by rewriting equation (2.187.1) as

$$\frac{dA}{dT} + (\lambda_A + \sigma_A\phi)A = \gamma_A\Sigma_f\phi,$$

or

$$\frac{dA}{dT} + \lambda_A{}^*A = \gamma_A\Sigma_f\phi. \tag{2.189.1}$$

Upon multiplying both sides by the integrating factor $e^{\lambda_A{}^*T}$, assuming ϕ to be constant, and by dT, the left side becomes a perfect differential, equal to $d(Ae^{\lambda_A{}^*T})$, so that

$$d(Ae^{\lambda_A{}^*T}) = \gamma_A\Sigma_f\phi e^{\lambda_A{}^*T}\,dT. \tag{2.189.2}$$

If $\Sigma_f\phi$ is constant, as postulated, integration of equation (2.189.2) gives

$$A(T) = \frac{\gamma_A\Sigma_f\phi}{\lambda_A{}^*}(1 - e^{-\lambda_A{}^*T}) + A(0)e^{-\lambda_A{}^*T}, \tag{2.189.3}$$

where $A(T)$ and $A(0)$ represent the concentration of A at times T and zero, respectively.

2.190. An alternative form of this equation is obtainable by introducing equation (2.188.1) and recalling the definition of $\lambda_A{}^*$ given above; thus,

$$A(T) = A_0(1 - e^{-\lambda_A{}^*T}) + A(0)e^{-\lambda_A{}^*T}, \tag{2.190.1}$$

so that, as T increases, $A(T)$ approaches the equilibrium value A_0. If, as in the operation of a freshly charged reactor, $A(0)$ is zero, the last term in equations (2.189.3) and (2.190.1) can be eliminated; for example, equation (2.190.1) becomes

$$A(T) = A_0(1 - e^{-\lambda_A{}^*T}). \tag{2.190.2}$$

2.191. Turning next to the daughter element B, this is removed from the system, by radioactive decay at the rate $-\lambda_B B$ and by neutron capture at the rate $-\sigma_B\phi B$. It is formed at the rate $\lambda_A A$ by the decay of its parent A, and in addition it may be produced at the rate $\gamma_B\Sigma_f\phi$ as a direct fission fragment. The net rate of growth of B is therefore given by

$$\frac{dB}{dT} = -\lambda_B B - \sigma_B\phi B + \lambda_A A + \gamma_B\Sigma_f\phi, \tag{2.191.1}$$

and the equilibrium or steady-state concentration of B is

$$B_0 = \frac{\lambda_A A_0 + \gamma_B\Sigma_f\phi}{\lambda_B + \sigma_B\phi} = \frac{\lambda_A A_0 + \gamma_B\Sigma_f\phi}{\lambda_B{}^*}, \tag{2.191.2}$$

where, as before,

$$\lambda_B{}^* = \lambda_B + \sigma_B\phi,$$

if the neutron flux remains constant. It will be noted that the value of A in equation (2.191.2) has been taken as the equilibrium concentration, since a steady state of B implies a steady state of A. Upon introducing equation (2.188.1) into equation (2.191.2), it is found that

$$B_0 = \frac{\lambda_A}{\lambda_A{}^*\lambda_B{}^*}\gamma_A\Sigma_f\phi + \frac{\gamma_B\Sigma_f\phi}{\lambda_B{}^*}. \tag{2.191.3}$$

2.192. Rearrangement of equation (2.191.1) gives

$$\frac{dB}{dT} + \lambda_B{}^*B = \lambda_A A + \gamma_B\Sigma_f\phi,$$

and, upon multiplying both sides by the integration factor $e^{\lambda_B{}^*T}$ and by dT, the result, assuming constant neutron flux, is

$$d(Be^{\lambda_B{}^*T}) = (\lambda_A A + \gamma_B\Sigma_f\phi)e^{\lambda_B{}^*T}\,dT.$$

Integration between 0 and T, with $\Sigma_f\phi$ constant, then yields

$$B(T) = e^{-\lambda_B{}^*T}\left[\int_0^T (\lambda_A A + \gamma_B\Sigma_f\phi)e^{\lambda_B{}^*T}\,dT + B(0)\right]. \tag{2.192.1}$$

Utilizing equation (2.189.3) for A, actually $A(T)$, this becomes, in the special case of a freshly charged reactor, when $A(0)$ and $B(0)$ are zero,

$$B(T) = \frac{\lambda_A}{\lambda_A{}^*}\gamma_A\Sigma_f\phi\left(\frac{1 - e^{-\lambda_B{}^*T}}{\lambda_B{}^*} + \frac{e^{-\lambda_B{}^*T} - e^{-\lambda_A{}^*T}}{\lambda_B{}^* - \lambda_A{}^*}\right) + \frac{\gamma_B\Sigma_f\phi}{\lambda_B{}^*}(1 - e^{-\lambda_B{}^*T}) \tag{2.192.2}$$

or, utilizing equations (2.188.1) and 2.191.2), for A_0 and B_0, respectively

$$B(T) = \frac{\lambda_A A_0}{\lambda_B{}^* - \lambda_A{}^*}(e^{-\lambda_B{}^*T} - e^{-\lambda_A{}^*T}) + B_0(1 - e^{-\lambda_B{}^*T}). \tag{2.192.3}$$

Here, again, as T increases, $B(T)$ approaches the equilibrium value B_0.

2.193. A case of interest, which arises in connection with poisoning by xenon-135, is when A has a very short life, especially in comparison with that of B, i.e., when λ_A is large. Then, $\lambda_A/\lambda_A{}^*$ is close to unity, and equation (2.191.3) reduces to

$$B_0 \approx \frac{(\gamma_A + \gamma_B)\Sigma_f\phi}{\lambda_B{}^*}. \tag{2.193.1}$$

In addition, A_0 will be small [cf. equation (2.188.1)], and so equation (2.192.3) becomes

$$B(T) \approx B_0(1 - e^{-\lambda_B{}^*T}). \tag{2.193.2}$$

Comparison of these expressions with equations (2.188.1) and (2.190.2), respectively, shows that, if the fission fragment A has a very short life, the daughter B may be treated as if it were itself a direct fission product formed with a yield of $\gamma_A + \gamma_B$. In this case, too, expressions of the form of equations (2.191.3) and (2.192.3) will give the values for C_0 and $C(T)$, respectively, except that all properties of A and B become those of B and C, respectively.

2.194. Another situation is that in which the daughter B, e.g., samarium-149, is a stable isotope, so that λ_B is zero. In these circumstances, equation (2.191.1) holds with elimination of the term $-\lambda_B B$, and the equilibrium concentration of

B is given by equation (2.191.2) as

$$B_0 = \frac{\lambda_A A_0 + \gamma_B \Sigma_f \phi}{\sigma_B \phi}. \tag{2.194.1}$$

This means that, provided σ_B is finite, that is to say, provided the species B has an appreciable cross section for neutron capture, it will attain an equilibrium concentration in the reactor in spite of the fact that it does not suffer radioactive decay. Further, since $\lambda_B{}^*$ is now equivalent to $\sigma_B\phi$, equation (2.192.3) becomes

$$B(T) = \frac{\lambda_A A_0}{\sigma_B \phi - \lambda_A{}^*} (e^{-\sigma_B \phi T} - e^{-\lambda_A{}^* T}) + B_0(1 - e^{-\sigma_B \phi T}).$$

2.195. Attention should be called to the fact that the existence of equilibrium or steady-state concentrations of A, B, etc., in all the cases considered above, depends on the postulate that both Σ_f and ϕ are constant. Although it is possible, in principle, to keep the flux constant in a reactor, the macroscopic fission cross section will decrease in the course of time. This is because Σ_f is equal to $N_f\sigma_f$, where N_f is the concentration of the nuclei undergoing fission, and this must decrease as the reactor operates. As a result, the so-called steady-state values, A_0, B_0, etc., decrease correspondingly. However, in all practical cases the fractional decrease in N_f is so small that it can be neglected for purposes of calculation.

Decay of Specific Fission Products After Shut-down

2.196. Suppose that, after operating for some time so that the steady state has been reached for A, B, etc., the reactor flux is decreased to a constant value ϕ'. It can be readily shown that equation (2.189.3) now becomes

$$A(t) = \frac{A\Sigma_f \phi'}{\lambda_A{}^*} (1 - e^{-\lambda_A{}^* t}) + A_0 e^{-\lambda_A{}^* t}, \tag{2.196.1}$$

where t is the time after the change was made in the flux, $\lambda_A{}^*$ is here equal to $\lambda_A + \sigma_A\phi'$, while A_0 is given by equation (2.188.1). If shut-down is complete, so that ϕ' is zero, the subsequent change in the concentration of A will be due only to radioactive decay; in this case, equation (2.196.1) becomes simply

$$A(t) = A_0 e^{-\lambda_A t}, \tag{2.196.2}$$

as expected [cf. equation (2.5.2)].

2.197. The value of $B(t)$, at time t after the flux has been reduced from ϕ to ϕ', is given by equation (2.192.1) with $\lambda_B{}^*$ equal to $\lambda_B + \sigma_B\phi'$, and ϕ replaced by ϕ'; the expression for $A(t)$ is now equation (2.196.1), and $B(0)$ is equal to B_0, the steady-state value before the flux was reduced. In the case of complete shut-down so that ϕ' becomes zero, equation (2.192.1) is transformed into

$$B(t) = e^{-\lambda_B t}\left[\int_0^t \lambda_A A e^{\lambda_B t}\, dt + B_0\right],$$

and, since $A(t)$ is now expressed by equation (2.196.2),

$$B(t) = e^{-\lambda_B t}\left[\int_0^t \lambda_A A_0 e^{(\lambda_B - \lambda_A)t}\, dt + B_0\right]$$

$$= \frac{\lambda_A}{\lambda_B - \lambda_A} A_0(e^{-\lambda_A t} - e^{-\lambda_B t}) + B_0 e^{-\lambda_B t}. \tag{2.197.1}$$

2.198. If $B(t)$, as given by equation (2.197.1), is plotted as a function of time t after complete shut-down, it is found that in certain circumstances $B(t)$ increases, then passes through a maximum, and finally decreases steadily (see Fig. 4.12). The physical interpretation of this fact is that, immediately after the reactor is shut down, and the neutron flux is virtually zero, the daughter B continues to be formed from A at about the same rate as before, and, although B decays, it is not now removed as a result of neutron capture. Consequently the amount of B in the system commences to increase. In due course, as A decays, the rate of formation of B decreases and may become smaller than its rate of radioactive decay. The concentration of B in the system thus ultimately falls off, after passing through a maximum. The application of these facts to reactor poisoning, when B is xenon-135, will be considered in Chapter IV.

2.199. The time after complete shut-down for attaining the maximum amount of B can be obtained by differentiating equation (2.197.1) with respect to t and setting the result equal to zero; thus,

$$\frac{dB(t)}{dt} = \frac{\lambda_A}{\lambda_B - \lambda_A} A_0(\lambda_B e^{-\lambda_B t} - \lambda_A e^{-\lambda_A t}) - B_0\lambda_B e^{-\lambda_B t} = 0$$

so that

$$t_{\max} = \frac{1}{\lambda_B - \lambda_A} \ln \frac{\lambda_B}{\lambda_A}\left[1 - \frac{\lambda_B - \lambda_A}{\lambda_A} \cdot \frac{B_0}{A_0}\right], \tag{2.199.1}$$

which is real, provided $\lambda_A > \lambda_B$. The value of $B_{\max}$ can then be obtained by substituting $t_{\max}$ for t in equation (2.197.1).

2.200. If B is a stable species, so that λ_B is zero, equation (2.197.1) for $B(t)$ after complete shut-down becomes

$$B(t) = A_0(1 - e^{-\lambda_A t}) + B_0,$$

the first term giving the amount of B formed by decay of A, with a concentration of A_0 at zero time, i.e., at shut-down, and the second term representing the amount of B at shut-down. Since B is stable, its concentration does not pass through a maximum but increases steadily after shut-down to a limiting value of $A_0 + B_0$, where B_0 is given by equation (2.194.1). The physical basis of this result will be readily apparent.

2.201. In the foregoing treatment it has been supposed, in order to avoid complicated expressions, that the reactor has essentially reached its steady state before shut-down. However, if this is not the case, the procedure is similar, except that the proper value of $A(T)$ or $B(T)$ attained in the reactor at shut-

down must be used for the initial concentration in estimating $A(t)$ and $B(t)$ after shut-down.

NEUTRON CAPTURE AND RADIOACTIVE DECAY PROCESSES

Formation of Neutron Capture Products

2.202. Various useful products, in particular radioisotopes and fissionable materials, such as plutonium-239, are formed by neutron capture in a nuclear reactor. It is of interest to derive expressions for the concentration of such products as a function of time. In the simplest case, suppose a stable species (absorber) is exposed in a reactor to a neutron flux ϕ and that it captures neutrons, forming A, which is radioactive and also captures neutrons. The expression for the variation in the concentration of A with time is identical with equation (2.187.1), except that the term $\gamma_A\Sigma_f\phi$ is replaced by $\Sigma\phi$, where Σ is the macroscopic capture cross section of the absorber. In this case, assuming Σ and ϕ to be constant, the equilibrium or steady-state concentration of A becomes

$$A_0 = \frac{\Sigma\phi}{\lambda_A^*}, \tag{2.202.1}$$

where λ_A^* has the same significance as before [equation (2.188.2)]. The expression for $A(T)$ is then given by equation (2.189.3) as

$$\begin{aligned} A(T) &= \frac{\Sigma\phi}{\lambda_A^*}(1 - e^{-\lambda_A^* T}) \\ &= A_0(1 - e^{-\lambda_A^* T}), \end{aligned} \tag{2.202.2}$$

the initial amount of A being taken as zero. It may be mentioned that the remarks in § 2.195 concerning the steady-state concentration apply equally here.

2.203. If, after a certain exposure, the product A is removed from the reactor, it undergoes normal radioactive decay, and its concentration $A(t)$ at time t after removal is given by

$$\begin{aligned} A(t) &= A(T)e^{-\lambda_A t} \\ &= A_0(1 - e^{-\lambda_A^* T})e^{-\lambda_A t}, \end{aligned} \tag{2.203.1}$$

where T is the time of exposure in the reactor. For a long exposure, so that the steady-state concentration of A has been attained,

$$A(t) = A_0 e^{-\lambda_A t}, \tag{2.203.2}$$

where A_0 is defined by equation (2.202.1). It will be shown in Chapter V how the foregoing equations may be used to determine neutron flux, e.g., in a reactor, by exposing a suitable absorber of known cross section and measuring the radioactivity of the product of the (n, γ) reaction. Alternatively, if the flux is known the same procedure can be used to evaluate the cross section of the absorber.

2.204. If it is required to determine $B(T)$, where B is formed when A captures a neutron, the methods used above for fission products can be readily adapted. As before, $\gamma_A\Sigma_f\phi$ is replaced by $\Sigma\phi$ and $\gamma_B\Sigma_f\phi$ is zero, since B is not formed directly from the original absorber, but only as a result of neutron capture by A. Hence, with these modifications, equations (2.191.2) and (2.202.1) yield

$$B_0 = \frac{\lambda_A}{\lambda_B^*} A_0 = \frac{\lambda_A}{\lambda_A^*\lambda_B^*}\Sigma\phi, \tag{2.204.1}$$

and from equation (2.192.2)

$$B(T) = \frac{\lambda_A\Sigma\phi}{\lambda_A^*}\left[\frac{(1 - e^{-\lambda_B^* T})}{\lambda_B^*} + \frac{e^{-\lambda_B^* T} - e^{-\lambda_A^* T}}{\lambda_B^* - \lambda_A^*}\right]. \tag{2.204.2}$$

The change in the concentration of B due to radioactive decay, after removal from the reactor, may be treated in a manner similar to that used for reactor shut-down.

2.205. When B decays it forms C, and this may or may not be radioactive. An important example of this type arises when uranium-238 captures a thermal neutron to form uranium-239, which represents A; the decay product B is then neptunium-239, and its daughter, i.e., C, is plutonium-239 (§ 1.59). Although the latter is radioactive, emitting an alpha particle, its half life is so long, i.e., its decay constant is so small, that for all practical purposes it may be regarded as being nonradioactive. Similar considerations apply to uranium-238, the initial neutron absorber.

2.206. Since the rate of formation of plutonium-239 in a production reactor (§ 1.136, *et seq.*) is of interest, this case will be considered. The net rate of increase of C, which is assumed to be nonradioactive but can absorb neutrons, is given by

$$\frac{dC}{dT} = \lambda_B B - \sigma_C\phi C, \tag{2.206.1}$$

where the first term on the right gives the rate of formation of C by decay of B, and the second term is the rate of loss due to neutron capture; σ_C is the total microscopic absorption cross section of C for all neutron processes, e.g., fission and radiative capture.

2.207. The equilibrium concentration of C, for constant flux ϕ, is then

$$C_0 = \frac{\lambda_B B_0}{\sigma_C\phi},$$

since B must have attained its steady-state value. The product C is then being lost, in radiative capture and fission processes, just as fast as it is formed from B. Introducing equation (2.204.1) for B_0, the expression for C_0 becomes

$$C_0 = \frac{\lambda_A}{\lambda_A^*} \cdot \frac{\lambda_B}{\lambda_B^*} \cdot \frac{\Sigma}{\sigma_C}, \tag{2.207.1}$$

where Σ is the macroscopic (capture) cross section for the original absorber, i.e.,

uranium-238 in this case. It should be noted, as in § 2.195, that the occurrence of a true steady state for plutonium-239 depends on the assumption that the concentration of uranium-238 remains essentially constant, and that the flux is also constant.*

2.208. The attainment of an equilibrium (or steady-state) concentration of plutonium-239 is important in connection with the behavior of production reactors. It shows that, even if the problems of radiation instability and of fission-product poisoning, referred to in Chapter I, could be overcome, there is still a limit to the useful operating time of the reactor, since a point will be reached when the plutonium-239 is being lost, as a result of fission and radiative capture, as fast as it is being formed from uranium-238.

2.209. Upon rearranging equation (2.206.1) to

$$\frac{dC}{dT} + \sigma_C \phi C = \lambda_B B,$$

multiplying by $e^{\sigma_C \phi T}\, dT$ and integrating, the result is

$$C(T) = e^{-\sigma_C \phi T} \lambda_B \int_0^T B e^{\sigma_C \phi T}\, dT, \qquad (2.209.1)$$

the initial concentration of C being taken as zero. The value of B is given by $B(T)$, as expressed by equation (2.204.2), and this can be inserted into equation (2.209.1) and the required integration performed.† The resulting expression gives the concentration of C, e.g., plutonium-239, as a function of time of reactor operation before the steady state is attained.

2.210. The methods described above can be used to derive equations for the time dependence and the equilibrium values of the concentrations of various reactor products formed by neutron capture, fission, or radioactive decay, or by various combinations of these processes (see, for example, §§ 7.10, 7.12). In some instances the setting up of the appropriate differential equations and their solution is tedious and time consuming. Consequently, generalized methods have been developed whereby the necessary calculations can be greatly accelerated. Since the basis of the treatment is somewhat involved, it will be sufficient to refer the interested reader to published reports on the subject.‡

Symbols Used in Chapter II

A	mass number
B	binding energy of electron
barn	10^{-24} cm²/nucleus
e	base of natural logarithms

* Although equation (2.207.1) for C_0 does not appear to contain ϕ explicitly, it is actually included in $\lambda_A{}^*$ and $\lambda_B{}^*$.

† See also, L. Gold, *Nucleonics*, **11**, No. 4, 40 (1953).

‡ R. F. Van Wye and J. G. Beckerley, *Nucleonics*, **9**, No. 4, 17 (1951); H. C. Ott, *ibid.*, **10**, No. 11, 116 (1952); see also, W. B. Lewis, *ibid.*, **12**, No. 10, 30 (1954).

E	energy (in ev or Mev)
E_b	binding energy of neutron
E_c	critical energy (for fission)
E_γ	gamma-ray energy
g	weight in grams
I	radiation (or neutron) intensity
k	Boltzmann constant (8.6×10^{-5} ev/°C or 4.8×10^{-5} ev/°F)
m	mass (of neutron)
n	neutron
n	number of neutrons
n	neutron density (neutrons/cm³)
$n(E)$	number of neutrons of energy E per unit energy interval
nv	neutron flux (neutrons/(cm²)(sec))
N	number of nuclei or atoms (or per cm³)
N_a	Avogadro number (6.02×10^{23})
p	proton
P	reactor power (watts)
R	range of particles
R	nuclear radius
t	time
t	time after fission
t	time after shut-down or after removal from reactor
$t_{\frac{1}{2}}$	half life of radioisotope
t_m	mean life of radioisotope
T	time after reactor start-up
T	time in reactor
T_0	reactor operating period
T	absolute temperature
T_K	absolute temperature on Kelvin scale
T_R	absolute temperature on Rankine scale
v	velocity (cm/sec)
w	weight in pounds
x	thickness of absorber
Z	atomic number
α	alpha particle
γ	gamma-ray photon
ϕ	neutron flux (neutrons/(cm²)(sec))
λ	radioactive decay constant
λ	mean free path
μ	linear absorption coefficient (cm⁻¹)
ν	number of atoms of given kind in molecule
ρ	density (g/cm³)
σ	microscopic cross section (cm²/nucleus or barns)

σ_a	absorption cross section (fission + capture)
σ_c	capture cross section
σ_f	fission cross section
σ_s	scattering cross section
σ_t	total cross section
Σ	macroscopic cross section (cm^{-1})
$\Sigma(E)$	macroscopic cross section for energy E
θ	Compton scattering angle
τ	time after reactor start-up

PROBLEMS

1. Using the cross sections given in this chapter, verify the values of η for thermal neutrons in Table 1.7. Calculate η for enriched uranium containing (*a*) 1 per cent, (*b*) 2 per cent, and (*c*) 5 per cent of uranium-235. What conclusion can be drawn from the results?

2. The radioactive decay constant of antimony-124 is given as 0.0115 day^{-1}. How many years would be required for the activity of this isotope to decay to 0.1 per cent of the initial value? How many half lives does this represent?

3. A sample of radioactive iodine has an activity of 1 mc, due to iodine-126, half life 13.0 days. What weight of this radioisotope is present?

4. Estimate the distance in inches the alpha particles from plutonium-239 would travel, before being stopped, in (*a*) animal tissue, (*b*) aluminum, (*c*) uranium.

5. If the fission product cesium-141 has an atomic mass of 140.9472 and that of its beta-decay product (barium-141) is 140.9438, estimate the approximate range of the beta particles, emitted by the former, in (*a*) air, (*b*) concrete of density 3.5 g/cm^3, assuming there are no gamma rays.

6. Derive a general expression relating the tenth-value layer thickness for gamma rays to the linear absorption coefficient of a material. What thickness of concrete of density 3.5 g/cm^3 would be required to attenuate 2-Mev gamma radiation by a factor of 10^3?

7. In traversing a thickness of 2.5 cm of material of atomic weight 112 and density 5.6 g/cm^3, the intensity of a collimated beam of neutrons is decreased to 0.1 of its initial value. Calculate the total microscopic cross section in barns and estimate the absorption cross section.

8. A bismuth (density 10 g/cm^3) "window" is used in some experimental reactors to attenuate the gamma radiation without appreciably affecting the thermal neutrons. Compare the ratio of 2-Mev gamma rays to thermal neutrons incident upon an 8.5-in. thick window to the emergent ratio.

9. The most probable mode of fission of uranium-235 leads to nuclei with mass numbers 95 and 139. Identify the particular isotopes formed, assuming that the neutron/proton ratio is the same in both fragments. By comparing the mass numbers with those of stable isotopes of the same elements, account for the beta activity of the fission products.

10. If neutrons can be regarded as a monatomic gas, estimate the "neutron gas pressure" (in atmospheres) equivalent to a thermal flux of 10^{12} neutrons/(cm^2)(sec) at 25°C (77°F). Note that at this temperature and 1 atm pressure, 1 gram atomic weight of a monatomic gas would occupy 22.6 liters.

11. From the Maxwell-Boltzmann equation (2.106.2) show that kT is the kinetic energy of a particle corresponding to the most probable velocity per unit velocity interval.

12. A photoneutron source consists of an antimony rod, external area 5 cm^2, having an activity of 0.1 curie (due to antimony-124), surrounded by 30 grams of beryllium (density 1.8 g/cm^3). Assuming that every nuclear decay of the antimony-124 is accompanied by a gamma-ray photon which emerges from the rod and that the cross section for the (γ, n) reaction with beryllium is 0.0011 barn, estimate the rate of neutron production.

13. The capture cross section of sulfur for neutrons of 0.025 ev (actual) energy is 0.54 barn. Calculate the average cross section for thermal neutrons at 80°C, assuming the $1/v$ law to be obeyed.

14. A Water Boiler type thermal reactor, which operates at an average temperature of 80°C, contains 1450 grams of uranyl sulfate (UO_2SO_4), enriched to the extent of 93 nuclear per cent in uranium-235, dissolved in ordinary water. The total volume of the solution is 13.5 liters and its density is 1.09 g/cm^3. Calculate the average absorption mean free path for thermal neutrons in the reactor.

15. By means of equation (2.89.1), calculate the excitation energy of the uranium-239 nucleus formed when uranium-238 captures a thermal neutron.

16. The Brookhaven reactor and the Materials Testing Reactor both operate at a power level close to 28,000 kilowatts, but the average thermal fluxes are 2×10^{12} and 2×10^{14} neutrons/(cm^2)(sec), respectively. If the Brookhaven reactor contains 50 tons of natural uranium, how many kilograms of uranium-235 are present in the core of the MTR?

17. A reactor operates at 30,000 kw for 100 days and is then shut down. What is the rate of energy release due to fission-product decay after a "cooling" period of 30 days? If the reactor contains 50 tons of natural uranium, what will be the activity in curies of a fuel element weighing 2.57 lb?

18. The mass of the O^{16} atom is 16.0000 and that of N^{16} is 16.0107 amu. Calculate (*a*) the minimum neutron energy in Mev necessary for the reaction $O^{16}(n, p)N^{16}$, and (*b*) the energy in Mev of the gamma rays accompanying the beta decay of N^{16} if the maximum beta-particle energy is 4.0 Mev.

19. How long would it require after removal of a natural-uranium fuel element from a reactor for 99 per cent of the uranium-239 still present to be converted to plutonium-239?

20. Cobalt-60, half life 5.3 yr, is made by exposing a 100-gram block of cobalt-59 (density 8.9 g/cm^3), capture cross section 37 barns, to thermal neutrons at a flux of 10^{14} neutrons/(cm^2)(sec). What length of exposure, in days, would be necessary to attain an activity of 1 curie? Determine the maximum (or equilibrium) activity of the cobalt-60 under the given conditions. The thermal-neutron absorption cross section of the latter may be taken to be 5 barns. (An excited state of cobalt-60 is formed to some extent, but as it decays to the ground state with a relatively short half life, it may be ignored.)

21. Tellurium-135 (half life 2 min) is formed in a fractional yield of 0.056 in the fission of uranium-235; this decays to iodine-135 (half life 6.7 hr) and the latter decays to xenon-135 (half life 9.2 hr), which is also formed directly to the extent of 0.003 in fission. Trace the growth of iodine-135 and xenon-135 with time in a reactor operating at a thermal flux of 2×10^{14} neutrons/(cm^2)(sec) and determine the equilibrium amounts relative to the number of fissionable nuclei present, the latter being assumed to remain essentially constant. The thermal-neutron capture cross section for iodine-135 is 7 barns and that for xenon-135 is 3.5×10^6 barns. (Note that because of its relatively short half life the tellurium-135 may be ignored and the assumption made that iodine-135 is produced directly to the extent of 0.056 in fission.)

Chapter III *

REACTOR THEORY: THE STEADY STATE

INTRODUCTION

3.1. This chapter is mainly concerned with the derivation of mathematical relationships between the critical size of a nuclear reactor and the properties of the materials of which it is constructed. The treatment is based upon the principle of the conservation or balance of neutrons, outlined in Chapter I. In any volume element of the reactor, the rate of change of neutron density with time is equal to the rate of production minus the rate of absorption and the rate of leakage. The general equation representing the neutron balance is, therefore,

$$\text{Production} - \text{Absorption} - \text{Leakage} = \frac{\partial n}{\partial t}, \tag{3.1.1}$$

where n is the neutron density and $\partial n/\partial t$ is its rate of change with time. When the system is at equilibrium, i.e., in a steady state, $\partial n/\partial t$ is zero; hence the steady-state equation is

$$\text{Production} = \text{Absorption} + \text{Leakage}. \tag{3.1.2}$$

3.2. If the reactor contains no extraneous neutron source, i.e., if the production of neutrons is due entirely to fission, then the steady state is equivalent to the critical state, and equation (3.1.2) is, in a sense, the critical equation. This result is, of course, identical with that given in § 1.94, namely, that in a critical system the effective multiplication factor, i.e., the ratio of the number of neutrons produced to the total number lost by absorption and leakage, is unity.

3.3. The problem of obtaining the conditions for reactor criticality reduces itself to that of deriving expressions for the respective rates, i.e., of production, absorption, and leakage, in equation (3.1.2). The rate of absorption depends upon various absorption cross sections, which must be determined experimentally, and the rate of production of neutrons by fission is also obtained from empirical considerations. Thus, if the nuclear properties of the multiplying system are known, the calculation of the production and absorption terms does not present any special difficulties.

* Reviewed by M. C. Edlund.

3.4. The determination of leakage, on the other hand, is not quite such a simple matter, although, it is of interest to note, the problem is one of classical mechanics rather than of nuclear physics. Leakage arises from the fact that neutrons are in motion; consequently more neutrons will, on the average, leave the reactor, where the neutron density (or concentration) is high, than will return to it from the surrounding space, where the density is low. In actual practice the neutron motion is affected by collisions with atomic nuclei which cause scattering to occur; this plays a major role in neutron diffusion upon which the calculation of leakage is based.

3.5. Neutrons undergo elastic scattering collisions with all nuclei and, as a result, a typical neutron trajectory consists of a number of straight path elements (see Fig. 3.1) joining the points (nuclei) where the neutron suffered scattering collisions. These are the scattering free paths of which the average is the scattering mean free path (cf. § 2.122). After a scattering collision the direction in which the neutron travels is not known exactly, but it can be given in terms of a probability distribution. However, when a large number of neutrons are considered, there is always a net movement of neutrons from regions of higher to those of lower neutron density. The rate at which this motion, referred to as *diffusion*, occurs can then be determined, to a first approximation, by means of simple and familiar laws.

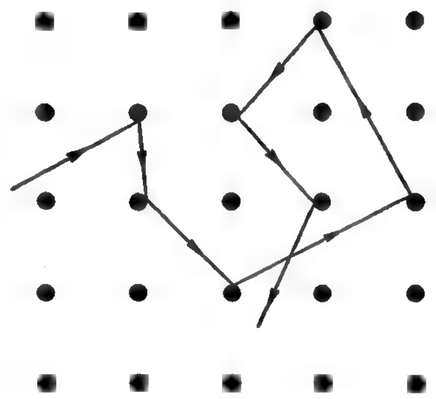


FIG. 3.1. Elastic scattering of neutrons by nuclei in a solid medium

3.6. In a reactor, the neutrons produced in fission are fast, and in an intermediate or thermal reactor they are subsequently slowed down by collisions with the nuclei of a moderator. During the slowing down process, the neutrons may suffer resonance capture or they may leak out. In a thermal reactor, the thermal neutrons diffuse for a time until they are either absorbed or escape. For purposes of theoretical treatment it is convenient to divide the problem into a number of parts.

3.7. The diffusion of monoenergetic neutrons, for which elastic scattering occurs without net energy loss, is discussed first. This represents a good approximation to the behavior of thermal neutrons in a weakly absorbing medium, where the Maxwell-Boltzmann distribution is maintained (§ 2.106). Next a general outline of the mechanics of elastic collisions will be given, so as to determine the energy loss suffered in a scattering collision. Then the problem of slowing down in a medium in which fast neutrons are being produced continuously will be divided into two parts. In the first place, consideration will be given to the distribution of neutrons as a function of energy, irrespective of their position. Subsequently, the spatial distribution of the neutrons, as a

result of diffusion during slowing down, will be considered. By combining the results, the characteristic equation for a critical reactor will be derived.

THE DIFFUSION OF NEUTRONS

Transport Theory

3.8. Neutron diffusion is, in some aspects, simpler than gaseous diffusion. Because of the extremely small density of neutrons in nuclear reactors, collisions between neutrons are rare.* Virtually all collisions leading to diffusion are consequently those between neutrons and the essentially stationary nuclei of the medium. In this respect, therefore, the phenomenon of neutron diffusion bears a greater resemblance to the diffusion of electrons in a metal than to gaseous diffusion. The rigorous treatment of neutron diffusion is, nevertheless, quite analogous to that used by Boltzmann in his classical studies of gaseous diffusion.

3.9. The method of approach is to consider a small volume element located at a certain point in the system, and to derive expressions for the various ways in which neutrons having a given velocity and moving in a specified direction, i.e., having a given velocity vector, enter and leave this volume element. For the steady state the algebraic sum of these expressions is equated to zero, and there is then obtained a complicated integro-differential equation, usually referred to as the (Boltzmann) transport equation. Even if detailed cross sections appearing in this equation were available, a complete solution would be prohibitively difficult and tedious in most cases. Therefore, certain approximations are made which lead to considerable simplification.

3.10. A reactor is a complex system, involving fuel material, which may consist only partly of fissionable elements, cladding of the fuel elements, coolant, structure, etc. It is thus almost impossible to describe the nuclear and other properties completely. Criticality calculations cannot be relied upon, therefore, to give more than an approximate indication of what is to be expected, but the results can be used as a guide to the design of experiments from which more precise data can be obtained.

Diffusion Theory Approximation

3.11. In the treatment of the transport equation, a simplification is achieved by supposing that all the neutrons have the same velocity (or energy) and that scattering collisions with nuclei do not involve any change of energy. These conditions apply, with a good degree of approximation, to neutrons in thermal equilibrium with a weakly absorbing medium, provided the cross sections used are properly averaged values (§ 2.128). Further, if the angular distribution of the neutron velocity vectors is independent of position, as it probably will be in regions that are more than about two scattering mean free paths distant from

* For the equivalent "neutron gas pressure," see Chapter II, Problem 10.

sources, strong absorbers, or boundaries, the transport equation for monoenergetic neutrons, which are scattered without energy loss, reduces to the so-called *diffusion theory approximation*. The results are widely used in the theoretical treatment of thermal reactors.

3.12. According to the diffusion theory approximation, as applied to neutrons, the quantity whose gradient determines the net rate of neutron flow is the neutron density. The familiar Fick's law of diffusion, for monoenergetic neutrons, can consequently be written as

$$\mathbf{J} = -D_0 \operatorname{grad} n, \tag{3.12.1}$$

where the neutron current vector $\mathbf{J}$ is the net number of neutrons flowing in a given direction in unit time through a unit area normal to the direction of flow; n, as before, is the neutron density, i.e., the number of neutrons per unit volume, at the point where $\mathbf{J}$ is observed, and D_0 is the diffusion coefficient.

3.13. For the subsequent treatment it is more convenient to use the neutron flux rather than the neutron density. Since the flux ϕ is equal to nv, where v is the (constant) neutron velocity, equation (3.12.1) can be modified to give

$$\mathbf{J} = -D \operatorname{grad} \phi, \tag{3.13.1}$$

where D is equal to D_0/v. The diffusion coefficient D for neutron flux, generally referred to merely as the *diffusion coefficient*, has the dimension of length; this follows from the fact that $\mathbf{J}$ and ϕ have the same dimensions.*

3.14. For a medium in which the absorption of neutrons is relatively weak, it can be shown from transport theory that, under conditions that the diffusion approximation is valid, the diffusion coefficient is given by

$$D = \frac{1}{3\Sigma_s(1-\bar{\mu}_0)} = \frac{\lambda_s}{3(1-\bar{\mu}_0)}, \tag{3.14.1}$$

where Σ_s is the total macroscopic scattering cross section of the medium for the monoenergetic neutrons, λ_s is the scattering mean free path ($=1/\Sigma_s$), and $\bar{\mu}_0$ is the average cosine of the neutron scattering angle per collision in the laboratory system.† The quantity $1/\Sigma_s(1-\bar{\mu}_0)$ or $\lambda_s/(1-\bar{\mu}_0)$ is called the *transport mean free path* and is denoted by λ_t; thus,

$$\lambda_t \equiv \frac{1}{\Sigma_s(1-\bar{\mu}_0)} = \frac{\lambda_s}{1-\bar{\mu}_0} \tag{3.14.2}$$

so that, from equation (3.14.1),

$$D = \tfrac{1}{3}\lambda_t. \tag{3.14.3}$$

* Both neutron current ($\mathbf{J}$) and the flux (ϕ) are usually expressed as neutrons per cm² per sec; it should be noted, however, that the current is a vector, i.e., it involves direction, whereas the flux is a scalar quantity.

† For a description of neutron collision in the laboratory and center of mass systems, see § 8.40, *et seq.*

3.15. It may be noted that, if the scattering were isotropic, i.e., spherically symmetric, in the laboratory system, the average cosine, $\bar{\mu}_0$, of the scattering angle would be zero, and λ_s would be identical with λ_t. The factor $1/(1 - \bar{\mu}_0)$, which may be regarded as the correction for anisotropic scattering, represents the preferential scattering in the forward direction, i.e., in the initial direction of motion of the neutron before collision with a nucleus. In general, $\bar{\mu}_0$ decreases with increasing mass of the struck nucleus, i.e., the tendency toward anisotropic scattering decreases. Hence, for heavy nuclei, the factor $1/(1 - \bar{\mu}_0)$ approaches unity and the scattering and transport mean free paths are not very different.

3.16. Returning to equation (3.13.1), it follows that since the vector $\mathbf{J}$ gives the number of neutrons flowing through unit area per second, in a given direction, the divergence (div) of this vector will represent the net number of neutrons leaving unit volume per second. This quantity is the rate of neutron leakage at the given point in the system where the flux is ϕ; thus, according to diffusion theory,

$$\begin{aligned}\text{Neutron leakage per cm}^3 \text{ per sec} &= -\text{div } D \text{ grad } \phi \\ &= -D\nabla^2\phi, \end{aligned} \tag{3.16.1}$$

where ∇^2 is the Laplacian operator. This may be expressed in various coordinates as follows:

Rectangular coordinates

$$\nabla^2 \equiv \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} \tag{3.16.2}$$

Spherical coordinates

$$\nabla^2 \equiv \frac{\partial^2}{\partial r^2} + \frac{2}{r} \cdot \frac{\partial}{\partial r} + \frac{1}{r^2 \sin\theta} \cdot \frac{\partial}{\partial \theta}\left(\sin\theta \frac{\partial}{\partial \theta}\right) + \frac{1}{r^2 \sin^2\theta} \cdot \frac{\partial^2}{\partial \varphi^2} \tag{3.16.3}$$

Cylindrical coordinates

$$\nabla^2 \equiv \frac{\partial^2}{\partial r^2} + \frac{1}{r} \cdot \frac{\partial}{\partial r} + \frac{1}{r^2} \cdot \frac{\partial^2}{\partial \theta^2} + \frac{\partial^2}{\partial z^2}. \tag{3.16.4}$$

The Diffusion Equation

3.17. For convenience, equation (3.1.1) may be rewritten as

$$-\text{Leakage} - \text{Absorption} + \text{Production} = \frac{\partial n}{\partial t}, \tag{3.17.1}$$

where the various terms on the left refer to rates per cm³ per sec, and n is the neutron density expressed as neutrons per cm³. The first term on the left of equation (3.17.1) is given by equation (3.16.1) as $D\nabla^2\phi$, the two negative signs leading to a positive sign. The second term can be derived from equation (2.121.2) and is accordingly $\Sigma_a\phi$, where Σ_a is the macroscopic absorption cross section for the monoenergetic (or thermal) neutrons. Upon making these substitutions, equation (3.17.1) becomes

$$D\nabla^2\phi - \Sigma_a\phi + S = \frac{\partial n}{\partial t}, \tag{3.17.2}$$

where S, called the *source term*, is the rate of production of neutrons per cm³ per sec. This expression, generally referred to as the *diffusion equation*, is basic to thermal reactor theory. In view of its derivation, it is strictly applicable only to monoenergetic neutrons and then only at distances greater than two or three mean free paths from strong sources, absorbers, or boundaries (cf. § 3.11). It will be applied here to thermal neutrons in particular; this is justifiable, provided the medium does not absorb neutrons strongly.

Diffusion Equation in Nonmultiplying Medium

3.18. For a system in the steady state $\partial n/\partial t$ is zero, and so the diffusion equation then becomes

$$D\nabla^2\phi - \Sigma_a\phi + S = 0. \tag{3.18.1}$$

In a nonmultiplying medium, i.e., one containing no fissionable material, the source term is zero everywhere except at the position of the neutron source. The differential equation (3.18.1) can be solved, with $S = 0$ for points outside the source region, and then the proper boundary conditions are applied at the source, as will be illustrated below. If S is set equal to zero, the steady-state diffusion equation (3.18.1) reduces to the homogeneous form applicable anywhere except at the source; thus,

$$D\nabla^2\phi - \Sigma_a\phi = 0 \tag{3.18.2}$$

or

$$\nabla^2\phi - \kappa^2\phi = 0, \tag{3.18.3}$$

where

$$\kappa^2 \equiv \frac{\Sigma_a}{D}. \tag{3.18.4}$$

Since Σ_a has the dimensions of reciprocal length (§ 2.112) and D of length, κ is a reciprocal length.

Solution of the Diffusion Equation: Boundary Conditions

3.19. The solution of the diffusion equation (3.18.3) will give the space distribution of the neutron flux at a steady state in a nonmultiplying medium at points other than the source. Since the diffusion equation is a differential equation, it does not provide a complete representation of the physical situation. The general solution will contain arbitrary constants of integration, and, in order to determine the proper values for these constants, it is necessary to apply restrictions upon the permitted solutions in the form of boundary conditions derived from the physical nature of the problem. The number of these boundary con-

ditions must be sufficient to provide a unique solution, with no arbitrary constants. Some of the boundary conditions which are frequently used in the solution of neutron distribution problems are examined in the following paragraphs.

3.20. It is to be expected that at the boundary between two different media there should be continuity of both neutron flux and neutron current. Thus, assuming monoenergetic (or thermal) neutrons, one simple boundary condition is the following: *At an interface between two media, with different diffusion properties, the neutron flux will be the same in both media.* If the media are indicated by the symbols A and B, then

$$(\phi_A)_0 = (\phi_B)_0, \tag{3.20.1}$$

the zero subscript being used to indicate that the values apply at the interface.

3.21. Since the neutron current is a vector, the continuity condition requires continuity of each of its components. If the neutron depends only on a single Cartesian coordinate, x, the boundaries must be planes normal to the direction of x, and the current boundary condition takes the form: *At a plane interface between two media, with different diffusion properties, the net neutron currents, in the direction normal to the interface, are equal.* The neutron current in the x-direction is given by equation (3.13.1) as

$$J_x = -D\frac{d\phi}{dx}, \tag{3.21.1}$$

so that the condition for continuity of current at the boundary may be written as

$$-D_A\left(\frac{d\phi_A}{dx}\right)_0 = -D_B\left(\frac{d\phi_B}{dx}\right)_0. \tag{3.21.2}$$

This result will hold at a spherical or infinite cylindrical interface as well as at a plane, the x-direction being the one for which the flux is independent of angle.

3.22. It should be noted that equation (3.21.2) implies that diffusion theory can be used at the boundary between the two media. Although this is actually not so, it has nevertheless been shown from transport theory that the equation represents a reasonably good approximation to the exact boundary condition.

The Linear Extrapolation Distance

3.23. At the boundary between a diffusion medium and a vacuum different circumstances exist, since there is no scattering back of neutrons from the vacuum into the diffusion medium. In other words, there is a flow of neutrons in one direction only. The boundary condition is now as follows: *Near the boundary between a diffusion medium and a vacuum the neutron flux varies in such a manner that linear extrapolation would require the flux to vanish at a definite distance beyond the boundary.* In general, the net neutron current represents the balance between currents in the positive and negative directions; according

to diffusion theory these are given by

$$J_+ = \frac{\phi}{4} - \frac{D}{2}\cdot\frac{d\phi}{dx}$$

and

$$J_- = \frac{\phi}{4} + \frac{D}{2}\cdot\frac{d\phi}{dx}$$

for the components in the x-direction. The difference between these two components gives the net current, in agreement with equation (3.21.1). In the case under consideration, i.e., at the medium-vacuum boundary, the negative component of the neutron current is zero, so that

$$J_- = \frac{\phi_0}{4} + \frac{D}{2}\cdot\frac{d\phi_0}{dx} = 0. \tag{3.23.1}$$

The flux ϕ_0 at the boundary is positive, and so it follows from equation (3.23.1) that the slope $d\phi_0/dx$ of the flux distribution must be negative at the boundary, as indicated schematically in Fig. 3.2.

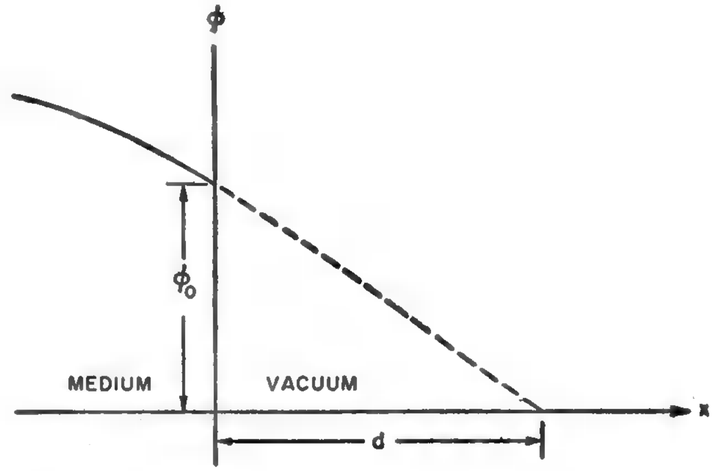


FIG. 3.2. Extrapolation of neutron flux at a plane interface between diffusion medium and a vacuum

3.24. If the neutron flux is extrapolated into the vacuum, using a straight line with the same slope as at the boundary, i.e., $d\phi_0/dx$, the flux would vanish at a distance d given by

$$-\frac{\phi_0}{d} = \frac{d\phi_0}{dx}.$$

Consequently, by equation (3.23.1),

$$d = 2D \tag{3.24.1}$$

or, since $D = \frac{1}{3}\lambda_t$, by equation (3.14.3),

$$d = \tfrac{2}{3}\lambda_t. \tag{3.24.2}$$

This distance is called the *linear extrapolation distance* or, sometimes, the *augmentation distance.* On the basis of linear extrapolation, therefore, the neutron flux should vanish at a distance of $\frac{2}{3}\lambda_t$ beyond the plane boundary between a diffusion medium and a vacuum. This boundary condition is sometimes stated in the form: the neutron flux vanishes at the extrapolated boundary, which lies $\frac{2}{3}\lambda_t$ (or $2D$) beyond the physical boundary in the case of a plane interface.

3.25. Since the diffusion theory approximation breaks down near a boundary, it is to be expected that the linear extrapolation distance obtained above is incorrect. According to the more exact transport theory treatment, the linear extrapolation distance at a plane surface of a weakly absorbing medium is $0.7104\lambda_t$, rather than $\frac{2}{3}\lambda_t$. In order to improve the results, therefore, the extrapolated boundary, when using diffusion theory, is taken to be $0.71\lambda_t$ beyond the actual plane boundary between a diffusion medium and a vacuum. The extrapolation distance is somewhat greater for curved than for plane surfaces. For a boundary of infinite curvature, i.e., of zero radius, this distance attains its maximum value of $\frac{4}{3}\lambda_t$.

3.26. It is important to realize, in postulating that the neutron flux vanishes at the extrapolated boundary, there is no implication, either from transport theory or diffusion theory, that the flux is actually zero there. The concept of the hypothetical boundary where the flux vanishes, as a result of linear extrapolation, is merely a convenient mathematical device used to obtain a simple boundary condition. Some indication of the actual state of affairs may be obtained with the aid of Fig. 3.3, which represents qualitatively the flux distribution in the x-direction near a medium-vacuum interface.

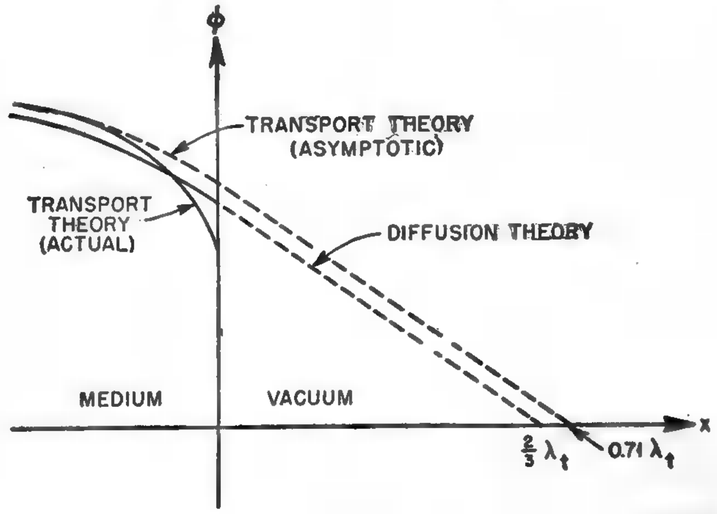


FIG. 3.3. Transport theory and diffusion theory extrapolations at a boundary

3.27. It is seen that the actual (or transport theory) flux undergoes a sharp decrease within a mean free path or so of the interface. Hence, in order that the boundary condition under consideration may be used in conjunction with diffusion theory to determine the neutron flux distribution at *reasonable distances from the interface*, a so-called asymptotic linear extrapolation of the transport theory distribution is made from the values not too near the boundary. The use of the resulting extrapolation distance of $0.71\lambda_t$ (or about $2.2D$) for a plane interface, together with diffusion theory, provides a satisfactory approximation for the behavior at points in the medium more distant than a transport mean free path from the interface.

Diffusion of Monoenergetic Neutrons from a Point Source

3.28. The procedure for solving the diffusion equation in a simple case will be illustrated by reference to the diffusion of monoenergetic neutrons from a point source into an infinite nonmultiplying medium. The result is of interest because it provides a physical interpretation of the quantity κ in equation (3.18.3) or, rather, of its reciprocal called the diffusion length (§ 3.34).

3.29. For convenience, the point source of neutrons, in the infinite medium, is assumed to be located at the origin of the coordinate system; the neutron distribution may then be regarded as having spherical symmetry. The Laplacian operator in equation (3.18.3) may thus be expressed in spherical coordinates with terms involving $d\theta$ and $d\varphi$ equal to zero; using equation (3.16.3), the source-free diffusion equation for the flux distribution becomes

$$\frac{d^2\phi}{dr^2} + \frac{2}{r}\cdot\frac{d\phi}{dr} - \kappa^2\phi = 0, \tag{3.29.1}$$

where r is the distance from the point source. This equation may be readily solved by setting $\phi \equiv y/r$, when it reduces to

$$\frac{d^2y}{dr^2} - \kappa^2 y = 0.$$

Since κ^2 is a positive quantity, as can be seen from equation (3.18.4), the general solution is

$$y = Ae^{-\kappa r} + Ce^{\kappa r},$$

and hence

$$\phi(r) = A\frac{e^{-\kappa r}}{r} + C\frac{e^{\kappa r}}{r}, \tag{3.29.2}$$

the constants A and C being determined by the boundary conditions.*

3.30. If C is not zero, then it is evident from equation (3.29.2) that the neutron flux would become infinite as $r \to \infty$. Since the flux must always be finite, it follows that C must be zero, so that the general solution of equation (3.29.1)

* Because B has a special significance in reactor theory (§ 3.108), it is not used here as an arbitrary constant.

applicable to the present case reduces to

$$\phi(r) = A\frac{e^{-\kappa r}}{r}. \tag{3.30.1}$$

3.31. In order to evaluate A use is made of what is called the source condition. If J is the neutron current density at the surface of a sphere of radius r, with the source at the center, then the total number of neutrons passing through the whole of the surface per second will be $4\pi r^2 J$. The limiting value of this number as r approaches zero will then be equal to the source strength Q neutrons/sec, i.e., to the number of neutrons emitted by the point source in all directions per second. The neutron current at a point distant r from the source is given by equations (3.13.1) and (3.30.1) as

$$J = -D\frac{d\phi}{dr} = DAe^{-\kappa r}\left(\frac{1+\kappa r}{r^2}\right).$$

Consequently, from the foregoing argument,

$$Q = \lim_{r\to 0} 4\pi r^2 J$$

$$= \lim_{r\to 0} 4\pi r^2 DAe^{-\kappa r}\left(\frac{1+\kappa r}{r^2}\right),$$

and hence

$$A = \frac{Q}{4\pi D}.$$

Equation (3.30.1) for the flux distribution accordingly becomes

$$\phi(r) = \frac{Q}{4\pi D}\cdot\frac{e^{-\kappa r}}{r}, \tag{3.31.1}$$

which is the required solution of the diffusion equation.

The Diffusion Length

3.32. From this result it is possible to derive an expression for the mean distance traveled by a neutron from its source to the point at which it is absorbed. However, instead of treating the first spatial moment of the flux, as would be required in this case, it is preferable, in view of later developments, to consider the second spatial moment. The volume of a spherical shell element of radius r and thickness dr surrounding the point source is $4\pi r^2 dr$. Since $\Sigma_a\phi$ gives the number of neutrons absorbed per cm³ per sec, in a region of flux ϕ and macroscopic absorption cross section Σ_a, it follows that the rate of neutron absorption in the spherical shell is $4\pi r^2 dr\Sigma_a\phi$ neutrons/sec. This is a measure of the probability that a neutron leaving the source, at the center of the shell, will be absorbed within the element dr at a distance r from the source.

3.33. Hence, the mean square (net vector) distance, $\overline{r^2}$, which a neutron travels from its source to where it is absorbed, is given by

$$\overline{r^2} = \frac{\int_0^\infty r^2(4\pi r^2\Sigma_a\phi)\,dr}{\int_0^\infty 4\pi r^2\Sigma_a\phi\,dr}.$$

Upon substituting the value for ϕ given by equation (3.31.1), this becomes

$$\overline{r^2} = \frac{\int_0^\infty r^3 e^{-\kappa r}\,dr}{\int_0^\infty re^{-\kappa r}\,dr} = \frac{6/\kappa^4}{1/\kappa^2} = \frac{6}{\kappa^2}. \tag{3.33.1}$$

3.34. A quantity called the *diffusion length*, represented by L, is defined by

$$L \equiv \sqrt{\frac{D}{\Sigma_a}} = \frac{1}{\kappa}, \tag{3.34.1}$$

bearing in mind the definition of κ as given by equation (3.18.4). It follows, therefore, from equation (3.33.1) that

$$L^2 = \tfrac{1}{6}\overline{r^2}, \tag{3.34.2}$$

and so the square of the diffusion length is equal to one sixth of the mean square (net vector) distance that a monoenergetic neutron travels from its source to the point at which it is absorbed by a nucleus.*

3.35. Although the foregoing derivations are applicable strictly to monoenergetic neutrons, they can be used for thermal neutrons in a weakly absorbing medium, provided properly averaged values of D and Σ_a are used (cf. § 2.128). In a reactor where the thermal neutrons are produced by the slowing down of fast neutrons, L^2 will be one sixth of the mean square (net vector) distance from the point at which a neutron becomes thermal to that at which it is absorbed.

3.36. The diffusion length of thermal neutrons in a nonmultiplying medium, such as a moderator (§ 1.56), can be determined experimentally by placing a rectangular block of the material over a uniform thermal-neutron source and then investigating, e.g., by the activation method (§ 2.203), the distribution of neutron flux within the block. Solution of the appropriate form of the diffusion equation gives the flux distribution in terms of κ^2 (or $1/L^2$) and the dimensions of the block. If the latter are known, the value of the diffusion length can then be calculated from the experimentally observed distribution.†

3.37. The measured values of the diffusion length, L, of thermal neutrons in four important moderating materials, namely, ordinary water, heavy water,‡ beryllium, and carbon (graphite), are recorded in Table 3.1. The macroscopic

* The net vector distance considered here differs from the absorption mean free path of the neutrons in the respect that the latter is the corresponding total scalar distance traveled by the neutron.

† For details of the theory and calculations, see S. Glasstone and M. C. Edlund, "The Elements of Nuclear Reactor Theory," D. Van Nostrand Co. Inc., 1952, § 5.67, *et seq.*

‡ It should be noted that the properties of heavy water given here and later in this chapter refer to the commercial product containing 99.75 per cent of deuterium oxide (D_2O).

TABLE 3.1. DIFFUSION PROPERTIES OF MODERATORS FOR THERMAL NEUTRONS

Moderator	Density (g/cm³)	L (cm)	Σ_a (cm^{-1})	D (cm)
Water	1.00	2.88	0.022	0.18
Heavy water (99.75% D_2O)	1.10	100	0.000085	0.85
Beryllium	1.84	23.6	0.0011	0.61
Carbon (graphite)	1.62	50.0	0.00037	0.92

absorption cross sections, Σ_a, for these substances are also given, as well as the diffusion coefficients, D, obtained from L and Σ_a by equation (3.34.1). The diffusion properties of a given material depend on its density and so this is quoted in each case. The transport mean free path λ_t is equal to $3D$, by equation (3.14.3), and the absorption mean free path λ_a is equal to $1/\Sigma_a$.

3.38. Some alternative relationships between the diffusion length and other properties of the medium are obtained by replacing D in equation (3.34.1) by $\frac{1}{3}\lambda_t$ and Σ_a by $1/\lambda_a$; the result is

$$L = \sqrt{\tfrac{1}{3}\lambda_t\lambda_a}.$$

For substances of high mass number λ_t is not very different from λ_s, as stated in § 3.15; then

$$L \approx \sqrt{\tfrac{1}{3}\lambda_s\lambda_a}$$

or, reconverting to macroscopic cross sections,

$$L \approx \frac{1}{\sqrt{3\Sigma_s\Sigma_a}}. \tag{3.38.1}$$

Infinite Plane Source in Medium of Finite Thickness

3.39. Another case in which the solution of the diffusion equation is of some interest is that in which monoenergetic (or thermal) neutrons from an infinite plane source diffuse into a nonmultiplying medium consisting of a slab of infinite extent but of finite thickness (Fig. 3.4). The situation here has some similarities to that of a reactor (neutron source) surrounded by a reflector (diffusion medium).

3.40. The coordinate system is chosen so that the source plane coincides with the plane for which $x = 0$ at all points. Since the source is assumed to be infinite in extent, it is evident that, for any given value of x, the neutron flux will be independent of y and z. The Laplacian operator in rectangular coordinates, from equation (3.16.2), is now merely d^2/dx^2; and so the diffusion equation (3.18.3) in the present case is

$$\frac{d^2\phi}{dx^2} - \kappa^2\phi = 0, \tag{3.40.1}$$

where, as before, κ is the reciprocal of the diffusion length in the medium. The general solution of equation (3.40.1), since κ^2 is a positive quantity, is

$$\phi(x) = Ae^{-\kappa x} + Ce^{\kappa x},$$

where A and C are constants to be determined by the boundary conditions.

3.41. In order to evaluate C use is made of the boundary condition that the flux shall vanish at the hypothetical extrapolated boundary (§ 3.23), whereas A

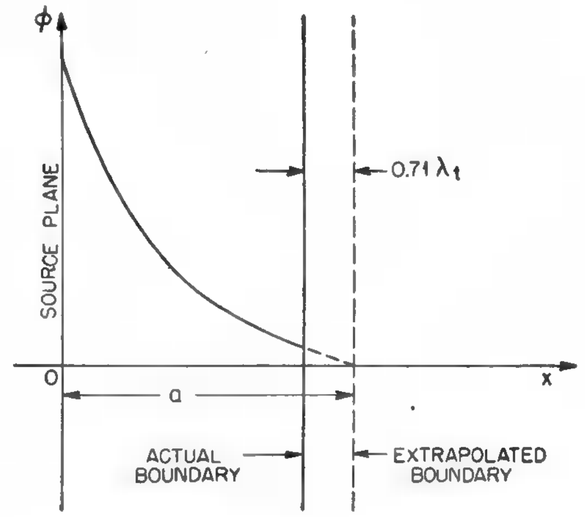


FIG. 3.4. Neutron flux distribution for plane source in finite medium

is derived from the source condition by a procedure analogous to that described in § 3.31. The result for the flux distribution in a medium of finite thickness, from a source of Q neutrons/(cm²)(sec), is

$$\phi(x) = \frac{\sinh\left[\kappa(a - x)\right]}{\kappa D \cosh(\kappa a)} Q, \tag{3.41.1}$$

where a is the thickness of the medium, *including the extrapolation distance.*

3.42. Some idea of the effect of the thickness of the diffusion medium on the flux distribution may be obtained from Fig. 3.5 in which ϕ/Q is plotted as a function of the distance x from a plane source in various thicknesses of graphite, namely, κa equal to 1, 2, 3, and infinity, respectively.* The value of $\kappa(=1/L)$ is 0.020 cm^{-1} and D is 0.92 cm (Table 3.1). Since κa is equal to a/L, it gives the thickness of the medium in terms of the diffusion length; thus the curves in Fig. 3.5 are for thicknesses of one, two, and three diffusion lengths and infinity.

3.43. It is seen that when κa is 3, i.e., when the extrapolated thickness of the medium is three times the diffusion length for the particular monoenergetic (or thermal) neutrons, the flux distribution of these neutrons is not greatly different

* Since equation (3.41.1) does not apply at the source itself, the curves are not extended to $x = 0$.

from that in an infinite medium, except near the boundary. Even when κa is 2, the difference from the infinite medium distribution is not large, at distances greater than a diffusion length from the boundary, i.e., for $x < 50$ cm in Fig. 3.5. These results are quite general, for κa, i.e., the thickness of the medium in terms of the diffusion length, is the important property in determining how the neutron flux falls off with distance from a plane source. Thus, when the thickness of a medium is two or more times the diffusion length, it may be treated as an infinite medium, at distances greater than a diffusion length or so from the boundary. This conclusion has an important bearing on the choice of thickness of the reflector for a reactor (§ 3.214). There is little to be gained by having the reflector more than two diffusion lengths in thickness.

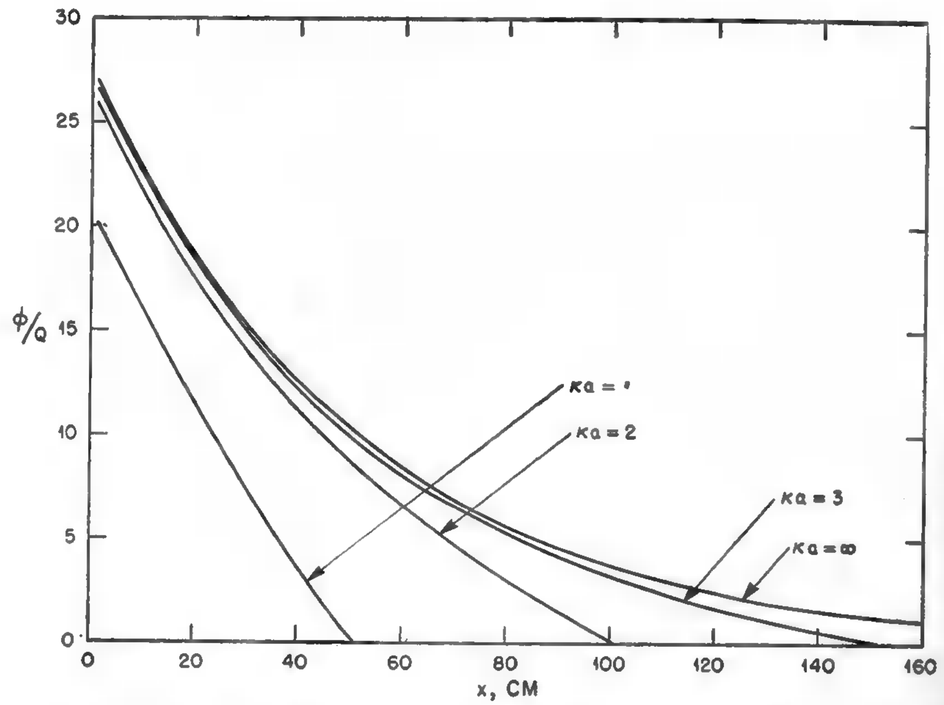


FIG. 3.5. Neutron flux distribution for media of different thicknesses

3.44. A consideration of the physical interpretation of these results is of interest. In the infinite medium there is no loss of neutrons, but in slabs of finite thickness neutrons will leak out at the boundary. If the thickness is three or more times the diffusion length, most of the neutrons are scattered back before they reach the boundary and the leakage is very small. For slabs of lesser thickness there is a greater loss of neutrons and consequently a more rapid falling off of the flux near the boundary. This effect is very marked when the thickness (including the extrapolation distance) is equal to the diffusion length, i.e., when $\kappa a = 1$, or less. A relatively small proportion only of the neutrons are scattered

back before they reach the boundary, and there is consequently a marked decrease in the flux as compared with media of greater thickness.

THE SLOWING DOWN OF NEUTRONS

The Mechanics of Elastic Scattering

3.45. The slowing down of fast neutrons is due almost entirely to elastic scattering collisions between the neutrons and the nuclei of the moderator.* Such collisions can be treated by the methods of classical mechanics, assuming the neutrons and scattering nuclei to behave as perfectly elastic spheres. By applying the principles of conservation of momentum and of energy, it is possible to derive a relationship between the scattering angle and the energy of the neutron before and after collision with a nucleus. Upon introducing an empirical scattering law, various useful results can be obtained.

3.46. In considering scattering collisions, two convenient frames of reference are used; these are the laboratory (L) system and the center of mass (C) system. In the former, the scattering nucleus is assumed to be at rest before the collision, whereas in the latter the center of mass of the neutron + nucleus is taken to be stationary in the collision. In the L system the viewpoint is essentially that of an external observer, but in the C system it is that of an observer who is located at the center of mass of the neutron + nucleus. For purposes of theoretical treatment the C system is simpler, although actual measurements are made in the L system.†

3.47. The conditions before and after a collision, as represented in the two systems, are shown in Fig. 3.6. The neutron, having a mass of unity on the conventional atomic mass scale, moves with a speed v_1 relative to the nucleus of mass number A. Since the target nucleus is stationary in the L system, the

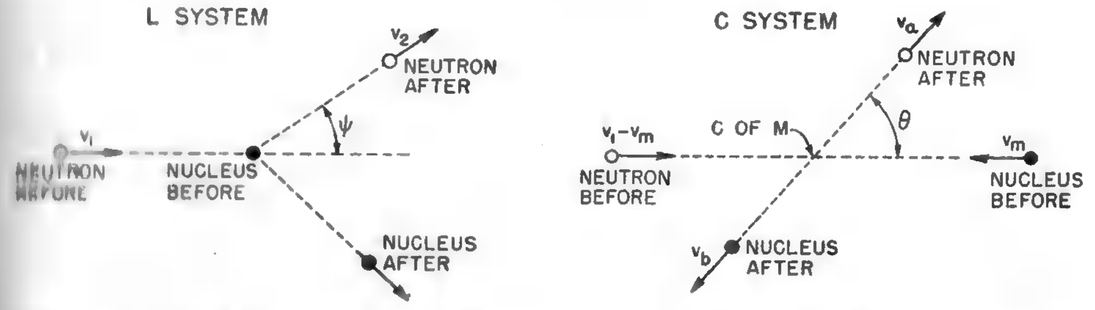


FIG. 3.6. Neutron scattering in laboratory (L) and center of mass (C) systems

* Inelastic scattering is neglected here since it is important only for neutron energies greater than about 0.1 Mev for elements of high mass number and about 1 Mev for those of low mass number (§ 2.100).

† This is true, provided the neutron energy is appreciably greater than the vibrational energy of the scattering atom; the nucleus may then be regarded as stationary with respect to the neutron.

momentum v_1 of the neutron is also the total momentum. The total mass of the colliding particles is $A + 1$, and consequently the speed v_m of the center of mass in the L system is given by

$$v_m = \frac{v_1}{A + 1}. \tag{3.47.1}$$

In the C system, the velocity of the neutron before collision is $v_1 - v_m$, and, using the value of v_m just derived, it follows that

$$v_1 - v_m = \frac{Av_1}{A + 1}. \tag{3.47.2}$$

3.48. It is seen, therefore, that in the C system the neutron and scattering nucleus appear to be moving toward one another with velocities of $Av_1/(A + 1)$ and $v_1/(A + 1)$, respectively. The momentum of the neutron (mass unity) is consequently $Av_1/(A + 1)$, while that of the nucleus (mass A) is also $Av_1/(A + 1)$, but in the opposite direction. The total momentum before collision, with respect to the center of mass, is then zero, and by the principle of the conservation of momentum it must also be zero after the collision.

3.49. Following the collision, the neutron in the C system leaves in the direction making an angle θ with its original direction; this is the scattering angle in the C system. The recoil nucleus must then move in the opposite direction, since the center of mass is always on the line joining the two particles. If v_a is the speed of the neutron and v_b that of the nucleus after collision, in the C system, then the requirement that the total momentum shall be zero is expressed by

$$v_a = Av_b. \tag{3.49.1}$$

3.50. The speeds of the neutron and nucleus before collision in the C system are given by equations (3.47.2) and (3.47.1), respectively, as seen above. The conservation of energy condition may therefore be written as

$$\frac{1}{2}\left(\frac{Av_1}{A + 1}\right)^2 + \frac{1}{2}A\left(\frac{v_1}{A + 1}\right)^2 = \frac{1}{2}v_a^2 + \frac{1}{2}Av_b^2, \tag{3.50.1}$$

where the left side gives the total kinetic energy before collision and the right side is that after collision. Upon solving equations (3.49.1) and (3.50.1) for v_a and v_b, it is found that

$$v_a = \frac{Av_1}{A + 1} \quad \text{and} \quad v_b = \frac{v_1}{A + 1}. \tag{3.50.2}$$

Comparison of these values with equations (3.47.2) and (3.47.1), respectively, shows that, in the C system, the speeds of the neutron and nucleus after collision are the same as the respective values before collision.

3.51. In order to determine the loss of kinetic energy of the neutron upon collision with an essentially stationary nucleus, it is necessary to transform the results obtained for the C system back to the L system. To perform this transformation, use is made of the fact that the two systems must always move relative to one another with the velocity (v_m) of the center of mass in the L system. Hence, the final velocity of the neutron after collision, in the L system, is obtained by adding the vector (v_m) representing the motion of the center of mass in the L system to the vector (v_a) indicating the velocity of the neutron after collision in the C system. This vector addition is shown in Fig. 3.7, the angle between the vectors being θ, the scattering angle in the C system.

3.52. If v_2 is the velocity of the neutron after collision in the L system, then by the law of cosines

$$v_2^2 = v_m^2 + v_a^2 + 2v_m v_a \cos\theta.$$

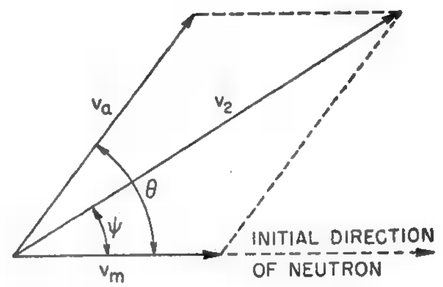


FIG. 3.7. Transformation from C to L systems

Introducing the values of v_m and v_a given by equations (3.47.1) and (3.50.2), respectively, the result is

$$v_2^2 = v_1^2 \frac{A^2 + 2A\cos\theta + 1}{(A + 1)^2}. \tag{3.52.1}$$

The angle ψ between the vectors v_2 and v_m is the scattering angle in the L system. It is the average value of the cosine of this angle for a large number of scatterings, i.e., $\overline{\cos\psi}$, which was represented by $\bar{\mu}_0$ in § 3.14.

ENERGY CHANGE IN SCATTERING

3.53. The kinetic energy E_1 of the neutron before a collision is $\frac{1}{2}mv_1^2$, and the energy E_2 after the collision is $\frac{1}{2}mv_2^2$; hence, the ratio of the neutron energy after collision to that before collision is given by equation (3.52.1) as

$$\frac{E_2}{E_1} = \frac{v_2^2}{v_1^2} = \frac{A^2 + 2A\cos\theta + 1}{(A + 1)^2}, \tag{3.53.1}$$

the assumption being made here, and subsequently, that the laboratory system, with an essentially stationary nucleus, represents the actual scattering conditions. If a quantity α, which is a property of the scattering nucleus related to its mass, is defined by

$$\alpha \equiv \left(\frac{A - 1}{A + 1}\right)^2 \tag{3.53.2}$$

then equation (3.53.1) becomes

$$\frac{E_2}{E_1} = \tfrac{1}{2}[(1 + \alpha) + (1 - \alpha)\cos\theta]. \tag{3.53.3}$$

3.54. The maximum value of the ratio E_2/E_1, i.e., the minimum loss of energy in a collision, occurs when $\theta = 0$, i.e., for a glancing collision; then $\cos\theta$ is unity,

and equation (3.53.3) gives

$$\frac{E_{\max}}{E_1} = 1 \quad \text{or} \quad E_{\max} = E_1. \tag{3.54.1}$$

In this event, the energies of the neutron before and after scattering are equal, and the neutron suffers no energy loss in the collision. The minimum value of E_2/E_1, i.e., the maximum possible energy transfer, occurs, on the other hand, when $\theta = \pi$, i.e., in a head-on collision. The value of $\cos\theta$ is now -1, and from equation (3.53.3),

$$\frac{E_{\min}}{E_1} = \alpha, \quad \text{or} \quad E_{\min} = \alpha E_1. \tag{3.54.2}$$

The minimum value of the energy to which a neutron can be reduced in an elastic scattering collision is therefore αE_1, where E_1 is the energy before the collision. The maximum fractional loss of energy in a collision is given by

$$\frac{E_1 - E_{\min}}{E_1} = 1 - \alpha, \tag{3.54.3}$$

and the actual maximum possible energy loss in a collision is $E_1(1 - \alpha)$.

3.55. Since, by equation (3.53.2), the quantity α is related to the mass number of the target nucleus, it is evident that the loss of energy suffered by a neutron in a collision will also depend on the mass number. For hydrogen, $A = 1$ and so $\alpha = 0$; it is, consequently, possible for a neutron to lose all of its kinetic energy in one collision with a hydrogen nucleus. This arises, of course, because the masses of the neutron and the hydrogen nucleus (proton) are essentially equal. For carbon $A = 12$ and $\alpha = 0.716$; hence, the maximum possible fractional loss of energy of a neutron in a collision with a carbon nucleus is $1 - 0.716 = 0.284$.

Empirical Scattering Law

3.56. Equation (3.53.3) gives the relationship between the energy ratio E_2/E_1 and the scattering angle θ in the C system, but derivation of an expression for the *average* energy change in a collision requires the introduction of a scattering law, i.e., a postulate concerning the probability of various scattering angles. Experiments indicate that the scattering of neutrons with energies of less than a few Mev is spherically symmetric, i.e., isotropic, in the C system. In other words, it appears from experiment that all values of $\cos\theta$ from -1 to $+1$ are equally probable. This empirical scattering law will be postulated as the basis of the subsequent treatment.

3.57. Although scattering may be spherically symmetric in the C system it will not be so in the L system unless the mass of the scattering nucleus is large compared to the mass of the neutron. If this is the case, the center of mass of the system is located virtually at the nucleus, and the L system becomes essentially identical with the C system. The same conclusion may be reached in another manner. It can be readily shown from Fig. 3.7 that

$$\cos\psi = \frac{A\cos\theta + 1}{\sqrt{A^2 + 2A\cos\theta + 1}}, \tag{3.57.1}$$

where ψ and θ are the scattering angles in the L and C systems respectively. For a heavy nucleus, $A \gg 1$, and then $\cos\psi \to \cos\theta$, so that the scattering angle in the L system approaches that in the C system. Therefore, if scattering is spherically symmetric in the latter, it will also be so in the former.

3.58. The average cosine of the scattering angle in the L system, i.e., $\bar{\mu}_0$, is given by

$$\bar{\mu}_0 = \frac{\int_0^{4\pi} \cos\psi \, d\Omega}{\int_0^{4\pi} d\Omega},$$

where $d\Omega$ is an element of solid angle. Transforming the variable Ω to θ, i.e., $d\Omega = 2\pi \sin\theta \, d\theta$, and introducing equation (3.57.1) for $\cos\psi$, based on isotropic scattering in the C system, it is found that

$$\bar{\mu}_0 = \frac{2}{3A}. \tag{3.58.1}$$

Thus $\bar{\mu}_0$ decreases with increasing mass of the scattering nucleus as stated earlier.

The Average Logarithmic Energy Decrement

3.59. Instead of calculating the average energy change in an elastic scattering collision, a more useful quantity in the study of the slowing down of neutrons is the average value of the decrease in the natural logarithm of the neutron energy per collision, or *the average logarithmic energy decrement per collision*. It is the average for all collisions of $\ln E_1 - \ln E_2$, i.e., of $\ln (E_1/E_2)$, where E_1 is the energy of the neutron before and E_2 is that after a collision. If this quantity is represented by the symbol ξ then, taking into consideration the equal probability of all values of $\cos\theta$ from -1 to 1, it follows that

$$\xi \equiv \overline{\ln \frac{E_1}{E_2}} = \frac{\int_{-1}^{1} \ln \frac{E_1}{E_2} \, d(\cos\theta)}{\int_{-1}^{1} d(\cos\theta)},$$

where the integration limits refer to $\cos\theta$. Upon substituting the expression for E_2/E_1 from equation (3.53.1), it is found that

$$\xi = 1 + \frac{(A-1)^2}{2A} \ln \frac{A-1}{A+1}$$

or, using the definition of α from equation (3.53.2),

$$\xi = 1 + \frac{\alpha}{1-\alpha} \ln \alpha. \tag{3.59.1}$$

For values of A in excess of 10, a good approximation to equation (3.59.1) may be written as

$$\xi \approx \frac{2}{A + \frac{2}{3}}. \tag{3.59.2}$$

Even for $A = 2$, the error of equation (3.59.2) is only 3.3 per cent.

3.60. It will be seen from equation (3.59.1) that the value of ξ is independent of the initial energy of the neutron, provided scattering is symmetrical in the C system.* In other words, in any collision with a given scattering nucleus, a neutron always loses, on the average, the same fraction of the energy it had before collision. This fraction decreases with increasing mass of the nucleus. It may be repeated here that the foregoing results involve the assumption of an essentially stationary nucleus prior to collision; this is justified only if the vibrational energy of the scattering nucleus is small compared to the kinetic energy of the neutron.

3.61. The values of α and ξ for a number of elements, especially those of low mass number, are given in Table 3.2. The average number of collisions with nuclei of a particular moderator required to decrease the energy of fission neutron, with initial energy of, say, 2 Mev, to the thermal value at ordinary temperatures, i.e., 0.025 ev, is obtained upon dividing $\ln (2 \times 10^6)/(0.025)$ by ξ for the given moderator; thus,

$$\text{Average number of collisions to thermalize (2 Mev to 0.025 ev)} = \frac{\ln\left(\frac{2 \times 10^6}{0.025}\right)}{\xi} = \frac{18.2}{\xi}. \tag{3.61.1}$$

Some of the results obtained from this equation are included in Table 3.2.†

TABLE 3.2. SCATTERING PROPERTIES OF NUCLEI

Element	Mass No.	α	ξ	Collisions to Thermalize
Hydrogen	1	0	1.000	18
Deuterium	2	0.111	0.725	25
Helium	4	0.225	0.425	43
Beryllium	9	0.640	0.206	86
Carbon	12	0.716	0.158	114
Uranium	238	0.983	0.00838	2172

* The arithmetic average value of E_1/E_2 per collision, which is equal to $\frac{1}{2}(\alpha + 1)$, is also independent of the initial neutron energy.

† The use of ξ to calculate the average number of collisions implies that the proper average value of E_1/E_2 to be employed in the calculation is the geometric average. This view is generally accepted, although it has been the subject of some controversy. If the arithmetic average of E_1/E_2, i.e., $\frac{1}{2}(1 + \alpha)$, is used, the collision numbers are somewhat different, especially for elements of very low mass number, e.g., 26 for H, 31 for D, and 119 for C.

Slowing Down Power and Moderating Ratio

3.62. According to equation (3.61.1), the value of ξ is inversely proportional to the number of scattering collisions required to slow down a neutron from fission energy to thermal values. It is thus a partial measure of the moderating ability of the scattering material. A good moderator is one in which there is a considerable decrease of neutron energy per collision on the average and, hence, it is desirable that ξ should be as large as possible. However, a large ξ is of little significance unless the probability of scattering, as indicated by the scattering cross section for neutrons with energy above thermal, is also large. The product $\xi\Sigma_s$, where Σ_s is the macroscopic scattering cross section of the moderator for epithermal neutrons, is called the *slowing down power*. It is a better measure than ξ alone of the efficiency of a moderator, for it is equal to ξ/λ_s, and so represents the average decrease in the logarithm of the neutron energy per cm of path. By equation (2.113.2), Σ_s is equal to $N_a\rho\sigma_s/A$, where N_a is the Avogadro number, ρ is the density of the moderator, σ_s its scattering cross section, and A the atomic (or molecular) weight; hence the slowing down power is expressed by $N_a\rho\sigma_s\xi/A$. The values for a number of materials consisting of (or containing) elements of low mass number are recorded in Table 3.3; the scattering cross sections are assumed to be constant in the energy range from 1 to 10^5 ev (cf. § 2.141).*

TABLE 3.3. SLOWING DOWN PROPERTIES OF MODERATORS

Moderator	*Slowing Down Power*	*Moderating Ratio*
Water	1.53 cm^{-1}	70
Heavy water	0.177	21,000
Helium*	1.6×10^{-5}	83
Beryllium	0.16	150
Carbon	0.063	170

* At atmospheric pressure and temperature.

3.63. Although the slowing down power gives a satisfactory indication of the ability of a material to slow down neutrons, it does not take into account the possibility that the substance may be a strong absorber of neutrons. The slowing down power of boron, for example, is better than that of carbon, but boron would be useless as a moderator because of its high cross section for neutron absorption (§ 2.138). In fact, any substance with appreciable absorption would be useless as a moderator. It is for this reason that boron and lithium are omitted from the tables given above; their slowing down power for neutrons is of no practical interest.

3.64. The ratio of the slowing down power, as defined above, to the macroscopic absorption cross section Σ_a, i.e., $\Sigma_s\xi/\Sigma_a$, called the *moderating ratio*, is

* For a compilation of data on moderators, see Table 8.9.

consequently an important quantity, from the theoretical standpoint, in expressing the effectiveness of a moderator. Some approximate values of this ratio, using absorption cross sections for thermal neutrons, are given in Table 3.3 above. It is seen that heavy water should be an excellent moderator. Where the employment of a liquid is not convenient, beryllium and carbon are evidently possible, but less efficient, alternatives to heavy water. Ordinary water can be used in certain circumstances, e.g., with enriched fuel, and has some advantages in respect to reactor size (§ 1.115). Helium under pressure, like water, could be used as both a moderator and heat-transfer medium, but this would involve structural difficulties. The choice of the moderator in any particular circumstances will thus depend on other factors, in addition to slowing down power and moderating ratio.

SLOWING DOWN IN INFINITE MEDIA WITHOUT ABSORPTION

The Collision Density

3.65. Suppose that fast neutrons are being produced at a uniformly definite rate, e.g., by fission, throughout the volume of a particular moderator. As the neutrons collide with the nuclei of the moderator, they steadily lose energy until they become thermal. But, since fast neutrons are continuously being generated, a steady-state distribution of neutron energies will soon be attained. This energy distribution will depend on the extent to which neutrons are absorbed in the system and on their escape from it while being slowed down. For the present, however, it will be postulated that the moderator is infinite in extent, so that there is no loss of neutrons by leakage, and also that there is no absorption while slowing down. The ultimate fate of the thermal neutrons is of no consequence at the moment.

3.66. If the source neutrons have energy E_0, assumed to be the same for all such neutrons, the minimum possible energy of a neutron after its first collision with a nucleus of moderator will be αE_0 (§ 3.54), where α is greater than zero, except for hydrogen (see Table 3.2). In the energy range between E_0 and αE_0, therefore, some of the neutrons will have suffered one collision, whereas others will have undergone two or more. The treatment is then somewhat complicated for moderators other than hydrogen, and so, in order to simplify it, the postulate will be made here that the neutron energies are all considerably less than αE_0, i.e., $E \ll \alpha E_0$. In other words, all the neutrons under consideration will have undergone many collisions, i.e., they have suffered multiple scattering, and there will be practically none that have been involved in only one collision.

3.67. Let $\phi(E')$ be the neutron flux per unit energy interval at E', and let $\Sigma_s(E')$ be the corresponding macroscopic scattering cross section. Then the number of scattering collisions per cm³ per sec experienced by those neutrons within an energy element dE' is $\Sigma_s(E')\phi(E')\,dE'$. This may be written in the form $F(E')\,dE'$, where $F(E')$, called the *collision density* per unit energy interval, is defined by

$$F(E') \equiv \Sigma_s(E')\phi(E'). \tag{3.67.1}$$

3.68. After collision with nuclei of moderator, the energy of the neutrons, initially E', will lie between E' and $\alpha E'$. The fraction of these neutrons scattered from E' into an energy element dE (Fig. 3.8) may be obtained in the following manner. If scattering is spherically symmetric in the C system, and if the L system is a good representation of the actual state of affairs, i.e., essentially stationary nuclei, then it can be shown that the fraction of neutrons scattered into an energy element dE is equal to dE divided by the maximum possible energy decrement per collision.* In the present case, the energy element is dE and the maximum possible energy decrement per collision for a neutron of energy E' is $E' - \alpha E' = E'(1 - \alpha)$; hence, the fraction required is $dE/E'(1 - \alpha)$.

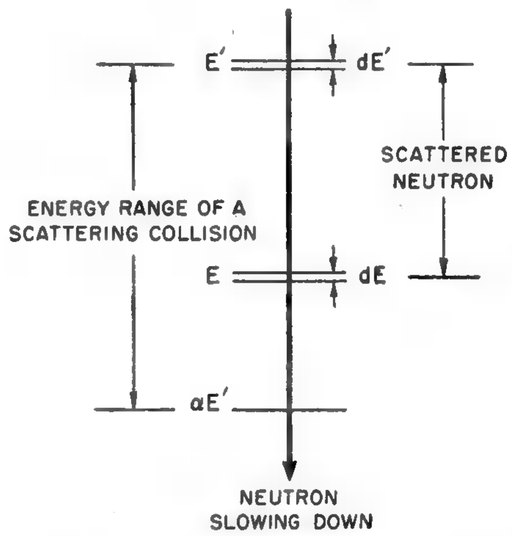


FIG. 3.8. Calculation of collision density and slowing down density in neutron scattering

3.69. The number of neutrons scattered per cm³ per sec into the energy element dE, from collisions in dE', is $F(E')\,dE' \times dE/E'(1 - \alpha)$, which is the total number of collisions in the element dE', i.e., $F(E')\,dE'$, multiplied by the fraction $dE/E'(1 - \alpha)$ that scatter neutrons into dE. The maximum initial energy of a neutron which could have energy E after such a collision is E/α. The total number of neutrons per cm³ per sec scattered into dE is thus obtained by integrating from E to E/α; that is,

$$\text{Number of neutrons per cm}^3\text{ per sec scattered into } dE = \int_E^{E/\alpha} \frac{F(E')\,dE'}{E'(1 - \alpha)}\,dE. \tag{3.69.1}$$

3.70. If $F(E)$ is the collision density per unit energy at E, the number of neutrons scattered out of the element dE is $F(E)\,dE$. Hence, in the steady state, when the number of neutrons scattered into dE must equal the number scattered out,

$$F(E) = \int_E^{E/\alpha} \frac{F(E')}{E'(1 - \alpha)}\,dE'. \tag{3.70.1}$$

A complete solution of this equation for all values of E less than αE_0 can be obtained, but, as stated above, the problem will be restricted to the so-called

* See Glasstone and Edlund, *op. cit.*, § 6.19.

asymptotic case, i.e., when $E \ll \alpha E_0$. In these circumstances, the solution of equation (3.70.1) is of the form

$$F(E) = \frac{C}{E}, \tag{3.70.2}$$

where C is a constant, as may be verified by substituting C/E' for $F(E')$ in equation (3.70.1) and performing the integration. This result applies, in general, when E is less than $\alpha^3 E_0$.

The Slowing Down Density

3.71. The problem now is to determine the value of the constant C in equation (3.70.2), and this is done by evaluating the *slowing down density*. This quantity is represented by the symbol q, and is defined as *the number of neutrons per cm³ per sec that slow down past a given energy E*. In the present case it may be obtained in the following manner. The fraction of collisions taking place in the energy element dE' which scatter past energy E is $(E - \alpha E')/E'(1 - \alpha)$. The numerator is the interval below E into which scattering from dE' (or energy E') is possible, and the denominator is the maximum energy range of neutrons scattered in the element dE' (cf. Fig. 3.8). Since $F(E')\,dE'$ is the total number of scattering collisions per cm³ per sec in the energy element dE', the number of these collisions leading to scattering past energy E is $F(E')\,dE' \times (E - \alpha E')/E'(1 - \alpha)$ per cm³ per sec. The total number of neutrons per cm³ per sec scattered past energy E, which is the slowing down density, is then obtained by integration between E and E/α, as in equation (3.69.1); thus,

$$q = \int_E^{E/\alpha} F(E') \frac{E - \alpha E'}{E'(1 - \alpha)}\, dE',$$

provided E is less than αE_0 so that no first collisions are involved.

3.72. Upon replacing $F(E')$ by C/E', according to equation (3.70.2), the expression for q, in the asymptotic case, becomes

$$\begin{aligned} q &= \frac{C}{1 - \alpha} \int_E^{E/\alpha} \frac{E - \alpha E'}{E'^2}\, dE' \\ &= C\left(1 + \frac{\alpha}{1 - \alpha} \ln \alpha\right) \\ &= C\xi, \end{aligned}$$

by equation (3.59.1) for ξ, based on isotropic scattering in the center of mass system.

3.73. If there is no leakage or absorption of neutrons, as was postulated in § 3.65, the same number of neutrons must slow down past each energy in the steady state, and this number must be equal to the source strength Q, i.e., the number of neutrons entering the system, at energy E_0, per cm³ per sec. Consequently for all energies

$$q = Q,$$

and hence the constant C is equal to Q/ξ. If this value is substituted into equation (3.70.2) the result, for the asymptotic energy region, is

$$F(E) = \frac{Q}{\xi E} = \frac{q}{\xi E}, \tag{3.73.1}$$

and from equation (3.67.1), which defines $F(E)$,

$$\phi(E) = \frac{Q}{\xi \Sigma_s E} = \frac{q}{\xi \Sigma_s E}. \tag{3.73.2}$$

As before, Σ_s is a function of the energy, although the argument is omitted here.

3.74. The results obtained above, such as equations (3.73.1) and (3.73.2), will apply to slowing down in any moderator for neutron energies less than about $\alpha^3 E_0$, e.g., $0.37E_0$ for carbon. It may be mentioned, however, that for hydrogen as moderator, for which ξ is unity and α is zero, the equations hold for neutrons of all energies.

3.75. If the moderator is a mixture or a compound, containing N nuclear species, the equations are still applicable, provided ξ is replaced by a mean value $\bar{\xi}$, defined by

$$\bar{\xi} = \frac{\Sigma_{s1}\xi_1 + \Sigma_{s2}\xi_2 + \cdots + \Sigma_{sN}\xi_N}{\Sigma_{s1} + \Sigma_{s2} + \cdots + \Sigma_{sN}}, \tag{3.75.1}$$

where the subscripts $1, 2, \ldots, N$ refer to the different nuclei. For water, for example, containing one hydrogen nucleus and two oxygen nuclei, the appropriate expression for $\bar{\xi}_{H_2O}$ is

$$\bar{\xi}_{H_2O} = \frac{\Sigma_{sH} + 2\Sigma_{sO}\xi_O}{\Sigma_{sH} + 2\Sigma_{sO}},$$

since ξ_H is unity.

SLOWING DOWN IN INFINITE MEDIA WITH ABSORPTION

The Resonance Escape Probability

3.76. The treatment of slowing down of neutrons in media in which absorption occurs is more difficult than that given above for nonabsorbing media. The only case which can be solved at all easily is that of moderation in a homogeneous mixture of hydrogen and a heavy absorber, such as uranium. However, an approximate solution is possible for the general case, of moderator with any mass number, which gives results that have been found useful.

3.77. Consideration will be given first to the *resonance escape probability;* this is represented by $p(E)$ and is defined as *the fraction of source neutrons which escape capture while being slowed down to a particular energy E*. For a thermal-neutron reactor, the energy of interest is, of course, the thermal value. The term "resonance" is used in describing the quantity $p(E)$ because most of the neutron captures will occur in the resonance region of the absorber, e.g., uranium-238.

If the resonance peaks are narrow and are widely spaced, the following non-rigorous procedure can be used to derive an expression for the resonance escape probability for any moderator-absorber system. It is assumed, as before, that the system is infinitely large, so that no neutrons escape from it.

3.78. Suppose the resonance energy region is divided into n intervals of widths, $\Delta E_1, \Delta E_2, \ldots, \Delta E_i, \ldots, \Delta E_n$. The probability that a neutron in slowing down will suffer collision with a nucleus in the energy interval ΔE_i is equal to $\Delta E_i/\xi E_i$, since $\Delta E_i/E_i$ (more correctly dE_i/E_i) is the width of the logarithmic interval corresponding to the energy interval ΔE_i, and ξ is the average change in ln E per collision in the medium under consideration.* For a homogeneous (uniform) system, the probability that the neutron will be absorbed in a collision is $\Sigma_a/(\Sigma_a + \Sigma_s)$, where Σ_a and Σ_s are the macroscopic absorption and scattering cross sections, respectively, for the system under consideration. The probability that a neutron will be absorbed in the energy interval E_i is thus given by

$$a_i = \frac{\Sigma_a}{\Sigma_a + \Sigma_s} \cdot \frac{\Delta E_i}{\xi E_i}, \tag{3.78.1}$$

and the probability, p_i, that the neutron will *escape* absorption in this interval is then

$$p_i = 1 - a_i.$$

Since a_i is, in general, much less than unity, this result may be written in the form

$$p_i \approx e^{-a_i}.$$

3.79. The over-all probability, p, that the neutron will escape capture in all n energy intervals is the product of the escape probabilities for the n individual intervals, so that

$$p \approx e^{-(a_1+a_2+\cdots+a_n)}, \tag{3.79.1}$$

where each of the a's is defined by the appropriate form of equation (3.78.1). If the energy intervals are made very small, the summation over the n intervals in the exponential term of equation (3.79.1) may be replaced by integration between energy E and the source energy E_0. The resulting p is then the resonance escape probability $p(E)$ at E, and by combining equations (3.78.1) and (3.79.1) this is seen to be

$$p(E) = \exp\left(-\frac{1}{\xi}\int_E^{E_0} \frac{\Sigma_a}{\Sigma_a + \Sigma_s} \cdot \frac{dE}{E}\right). \tag{3.79.2}$$

This result is frequently used to express the resonance escape probability in a homogeneous mixture of fuel and moderator.

3.80. For a mixture consisting of hydrogen as moderator and an absorber of high mass number, the value of ξ is close to unity. In these circumstances,

* In the general case, ξ is really the average value $\bar{\xi}$ defined by equation (3.75.1).

equation (3.79.2) becomes essentially identical with one derived in a more rigorous manner. Equation (3.79.2) may, therefore, be regarded as a fair approximation for all systems of interest.

3.81. An alternative definition of the resonance escape probability, equivalent to the one given in § 3.77, is the ratio of the slowing down density at energy E with absorption to the value without absorption, i.e.,

$$p(E) = \frac{q^*(E)}{q(E)}, \tag{3.81.1}$$

where $q^*(E)$ represents the slowing down density in an absorbing system.

3.82. A relationship between $q^*(E)$ and the flux $\phi(E)$, analogous to equation (3.73.2), can be derived, but the treatment is lengthy and will not be given here. A satisfactory result can, however, be obtained in a simple, although not exact, manner. If the collision density for neutrons of energy E in an absorbing medium is represented by $F^*(E)$, then, by analogy with equation (3.73.1), it should be possible to write

$$F^*(E) = \frac{q^*}{\xi E},$$

where the starred quantities refer to the behavior in an absorbing medium. Alternatively, by the arguments in § 3.67, the total collision density for scattering and absorption may be represented by a modified form of equation (3.67.1), i.e.,

$$F^*(E) = (\Sigma_a + \Sigma_s)\phi(E),$$

where $\Sigma_a\phi(E)$ is the contribution from absorbing collisions and $\Sigma_s\phi(E)$ is that from scattering collisions. Upon comparing these two expressions for $F^*(E)$, it is seen that

$$\phi(E) = \frac{q^*}{\xi(\Sigma_a + \Sigma_s)E} \tag{3.82.1}$$

or, introducing equation (3.81.1) for the resonance escape probability,

$$\phi(E) = \frac{pq}{\xi(\Sigma_a + \Sigma_s)E}, \tag{3.82.2}$$

where q is the value the slowing down density would have for neutrons of energy E if there were no absorption in the given medium.

SPATIAL DISTRIBUTION OF SLOWED DOWN NEUTRONS

Fermi Age (Continuous Slowing Down) Model

3.83. The next step in the discussion of the slowing down of neutrons is to investigate the spatial distribution of neutrons of various energies, resulting from diffusion during the slowing down process. The problem is important because the average (net vector) distance a fission neutron travels while slowing down

determines the neutron leakage in a finite medium, and so has a direct bearing on the critical size of a reactor. If this average distance is large, then the reactor will have to be large, so as to reduce the probability of escape of neutrons before they have been slowed down to thermal energies. Thus the distribution of neutrons in space while slowing down determines their loss by leakage.

3.84. A relatively simple, but approximate, approach to the problem, which is not applicable to media containing very light nuclei, e.g., hydrogen and deuterium, is based on the *continuous slowing down model*, sometimes called the *Fermi age model*. Consider a neutron produced at the fission energy E_0; it will travel for a certain time with this energy before it collides with a nucleus. After the collision its energy will be decreased, and it will then travel with this constant lower energy until it meets another nucleus. Since the neutron is now moving with a smaller speed, the time between collisions is, on the average, increased. This process of alternate movement at constant energy followed by a collision in which the energy is lowered will continue until the neutron is thermalized.

3.85. Since the average change in the logarithm of the neutron energy per collision, i.e., ξ, is independent of the energy, a plot of ln E of a neutron against time t from fission energy (E_0) down to thermal energy (E_{th}) should be of the form of Fig. 3.9. This consists of a series of steps of approximately equal height ξ, but of gradually increasing length. The horizontal lines represent the constant energy of the neutron while it is diffusing between collisions, and the lengths of these lines give the times elapsing between successive collisions.

3.86. Because individual neutrons behave differently, even if originating with the same energy and diffusing in the same medium, the plot of ln E against t, as in Fig. 3.9, will vary from one neutron to another. If the medium consists of nuclei of moderate or high mass number, the spread of energies among the neutrons of the same generation is relatively small, and it is possible to represent

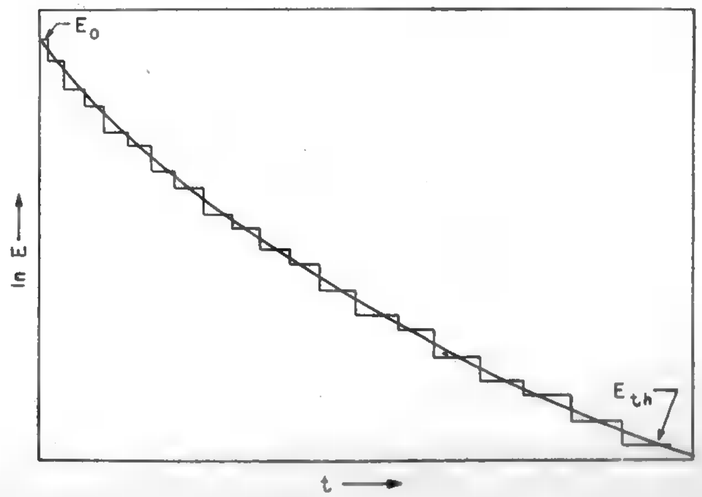


FIG. 3.9. Continuous slowing down approximation

the behavior of a large number of individual neutrons by means of an average. In these circumstances, too, the logarithmic change of energy per collision is small; that is to say, the height of the steps in Fig. 3.9 will be small. As a result, the series of steps may be replaced, without serious error, by a continuous curve, such as that shown in the diagram. This is the basis of the continuous slowing down (Fermi age) model. It is assumed that, during slowing down, neutrons lose energy continuously rather than discontinuously as is actually the case.

3.87. It should be emphasized that the continuous slowing down model can be valid, even as an approximation, only for moderators consisting of elements of fairly high mass number. It does not apply to hydrogen, deuterium, or to moderators containing these atoms, e.g., ordinary water or heavy water.* It is possible, as already seen, for a neutron to lose all its energy in a single collision with a hydrogen nucleus. There is, consequently, a considerable spread of energies and it is not justifiable to represent the slowing down characteristics of a large number of neutrons by an average behavior. Further, because of the relatively large changes in neutron energy per collision, and hence the small number of collisions required to reduce the energy to thermal values (see Table 3.2), the representation of the slowing down as continuous is completely invalid. The procedure used in connection with the slowing down distribution in water as moderator will be considered later (§ 3.162).

The Age Equation

3.88. The first problem is to develop an equation which will represent the spatial distribution of the slowing down density according to the continuous slowing down model in a nonabsorbing medium. The following derivation, although not completely rigorous, leads to the correct result in a relatively simple manner and indicates the general method of approach. Suppose that the neutrons are slowing down in a nonabsorbing medium, and after a time t they have a velocity v. Let λ_s be the scattering mean free path, i.e., the average distance a neutron of this velocity travels between successive collisions with nuclei. The number of collisions a neutron suffers during the subsequent time interval dt is then $v\,dt/\lambda_s$. Since ξ is the average logarithmic energy change per collision, it follows that the decrease in ln E is equal to ξ times the number of collisions in the time interval dt; hence

$$-d \ln E = \frac{v\xi}{\lambda_s}\,dt$$
$$= v\xi\Sigma_s\,dt,$$

where $-d$ ln E is the logarithmic energy loss during the time dt and Σ_s (equal to $1/\lambda_s$) is the macroscopic scattering cross section of the medium. Upon replacing d ln E by dE/E and rearranging, the result is

* The continuous slowing down model appears to be reasonably good even for heavy water (private communication from M. C. Edlund).

$$-\frac{dE}{dt} = v\xi\Sigma_s E, \tag{3.88.1}$$

where dE is the energy loss suffered by a neutron in the time interval dt, on the basis of the continuous slowing down model.

3.89. At a given point away from a source in a nonabsorbing medium, the general diffusion equation (3.17.2) for neutrons of energy E becomes

$$\frac{\partial n}{\partial t} = D\nabla^2\phi,$$

which means, in physical terms, that the rate at which the neutron density increases with time, at the given point, is equal to the net rate at which neutrons diffuse to that point. Since ϕ is equal to nv, this result may be written as

$$\frac{1}{v} \cdot \frac{\partial\phi(E)}{\partial t} = D\nabla^2\phi(E),$$

where, for convenience, the flux is represented by $\phi(E)$, which is defined in § 3.67 as the neutron flux of energy E per unit energy interval. Using the value of $\phi(E)$ given by equation (3.73.2), it follows that

$$\frac{1}{v\xi\Sigma_s E} \cdot \frac{\partial q}{\partial t} = \frac{D}{\xi\Sigma_s E}\nabla^2 q, \tag{3.89.1}$$

where q is the slowing down density for neutrons of energy E at the point under consideration. By combining the general mathematical relationship

$$\frac{\partial q}{\partial t} = \frac{\partial q}{\partial E} \cdot \frac{\partial E}{\partial t}$$

with equations (3.88.1) and (3.89.1), the result is

$$\nabla^2 q = -\frac{\xi\Sigma_s E}{D} \cdot \frac{\partial q}{\partial E}. \tag{3.89.2}$$

It should be noted that both Σ_s and D are functions of the neutron energy, although the argument is not included in equation (3.89.2).

3.90. A new variable $\tau(E)$ at energy E is now defined by

$$\tau(E) \equiv \int_E^{E_0} \frac{D}{\xi\Sigma_s E}\, dE, \tag{3.90.1}$$

where E_0 is the energy of the source neutrons. Upon making the transformation, whereby the variable E is replaced by τ, equation (3.89.2) reduces to

$$\nabla^2 q = \frac{\partial q}{\partial \tau}, \tag{3.90.2}$$

and this is known as the *Fermi age equation*. The quantity $\tau(E)$ is called the *Fermi age* or the *symbolic age*. It should be noted, however, that it is not a unit

of time, but rather a length squared. This can be readily seen from equation (3.90.2), when it is realized that the Laplacian operator implies differentiation twice with respect to distance. Thus equation (3.90.2) represents the spatial distribution of the slowing down density in a nonabsorbing medium.

3.91. Although the age does not represent an elapsed time, it is nevertheless related to the chronological age of the neutrons, i.e., the time elapsing between formation as a source (fission) neutron and the attainment of a given energy. This can be seen, in a general way, from equation (3.90.1) which defines the age. When $E = E_0$, i.e., for neutrons of source energy, τ is zero. As E decreases, however, it is seen that $\tau(E)$ increases correspondingly. In other words, as the neutron slows down its age increases.

3.92. The foregoing results apply to a medium in which there is no absorption of neutrons, but it can be shown that with a slight modification the age equation can be used for a weakly absorbing medium. It is found that if $q(E)$ is a solution of equation (3.90.2) for the case of no absorption, then the corresponding slowing down density $q^*(E)$, for the case with (weak) absorption, is given, to a sufficient approximation, by equation (3.81.1), i.e.,

$$q^*(E) = p(E)q(E), \tag{3.92.1}$$

where $p(E)$ is the resonance escape probability for neutrons of energy E. In general, therefore, at any energy, the slowing down density with absorption that satisfies the Fermi age equation may be taken as equal to the slowing down density solution of the Fermi equation without absorption, at the same energy, multiplied by the resonance escape probability for that energy.

Solution of the Age Equation: Significance of Age

3.93. The Fermi age equation (3.90.2) is of a form that is familiar to engineers; it is the same, for example, as the equation representing the transient conduction of heat in a continuous medium containing no sources.* Solutions for various forms are thus to be found in the literature. A case of immediate interest is that of a point source of fast neutrons, assumed to be monoenergetic, undergoing continuous slowing down in an infinite nonabsorbing medium. The solution of the age equation is then

$$q(r, \tau) = \frac{e^{-r^2/4\tau}}{(4\pi\tau)^{\frac{3}{2}}}, \tag{3.93.1}$$

* The general form of the heat-conduction equation is

$$\nabla^2 T = \frac{1}{\alpha} \cdot \frac{\partial T}{\partial t},$$

where T is the temperature, t the time, and α is the thermal diffusivity (see W. H. McAdams, "Heat Transmission," 3rd ed., McGraw-Hill Book Co. Inc., 1954, page 34). It will be noted that τ in the Fermi age equation actually corresponds to a time in the heat-conduction equation, the difference in dimensions being accounted for by α which has the dimensions of (length)^2/time.

where $q(r, \tau)$ is the slowing down density for neutrons of age τ at a point distant r from the source. This expression is that for a Gauss error curve, and, consequently, the distribution of slowing down density for a given age is sometimes referred to as Gaussian.

3.94. The shape of the curve representing the variation of $q(r, \tau)$ with r, for a given value of τ, is shown in Fig. 3.10. It is seen that when τ is small, i.e., the neutron is near the source energy, the curve is high and narrow. For τ large, i.e., for neutrons of lower energy, however, the curve is lower and more spread out. This is to be expected on physical grounds. A small value of the age means that the neutrons have suffered little slowing down, and so they have not

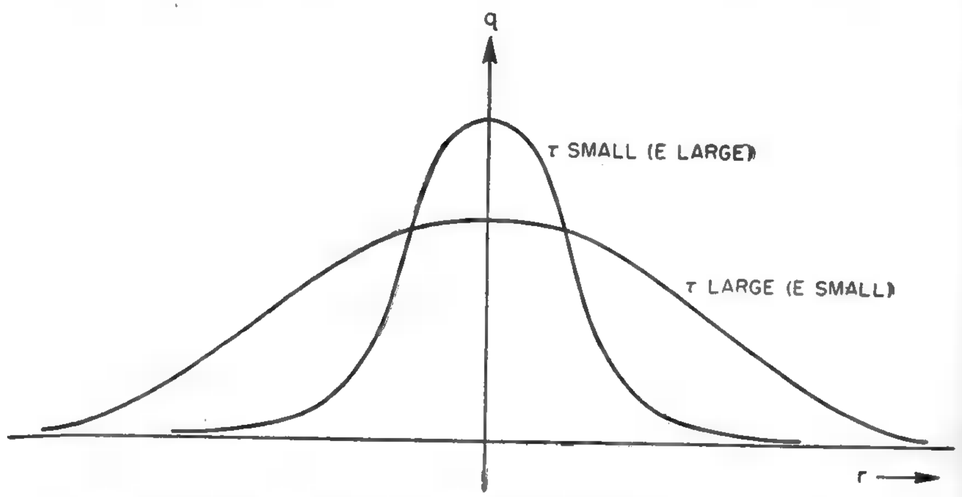


FIG. 3.10. Slowing down density distribution about point source

diffused far from the source. Consequently most of the neutrons will have high energies and be in the neighborhood of the source; this corresponds to a high and narrow distribution curve. On the other hand, when τ is large, the neutrons will have undergone considerable slowing down and will have diffused appreciable distances from the source. The distribution curve of the slowing down density will thus be low and broad.

3.95. A more precise physical significance can be associated with the Fermi age by calculating the mean square (net vector) slowing down distance, denoted by $\overline{r_s^2}$, about a point source, in a manner similar to that used in connection with the diffusion length (§ 3.33). Thus,

$$\overline{r_s^2} = \frac{\int_0^\infty r^2[4\pi r^2 q(r, \tau)]\, dr}{\int_0^\infty 4\pi r^2 q(r, \tau)\, dr},$$

where $q(r, \tau)$ is given by equation (3.93.1), so that

$$\overline{r_s^2} = \frac{\int_0^\infty r^4 e^{-r^2/4\tau}\, dr}{\int_0^\infty r^2 e^{-r^2/4\tau}\, dr} = 6\tau$$

or

$$\tau = \tfrac{1}{6}\overline{r_s^2}. \qquad (3.95.1)$$

The age τ is consequently one sixth of the mean square (net vector) distance traveled by a neutron from the time of its emission from the source, i.e., age zero, to the time at which its age is τ.*

3.96. The age of neutrons of specified energy in a given medium can be determined experimentally, provided a material is available which strongly absorbs neutrons of that energy. For example, the slowing down density can be measured at indium resonance, i.e., energy about 1.4 ev, at various distances from a point source of fast neutrons. Upon taking logarithms of equation (3.93.1), it is seen that

$$\ln q(r) = \text{const.} - \frac{r^2}{4\tau},$$

so that the plot of $\ln q$ against r^2 should be a straight line with a slope of $-1/4\tau$. This will give the age of indium resonance neutrons, and a correction can be applied to obtain the age of thermal neutrons, if required. Some values of the age of thermal neutrons at 20°C from fission sources in four important moderating media are given in Table 3.4.

TABLE 3.4. AGE OF THERMAL NEUTRONS AT 20°C FROM FISSION SOURCE

Moderator	*Age* (cm²)
Water	33
Heavy water	120
Beryllium	98
Graphite	350

3.97. It will be noted that Table 3.4 contains data for the age in ordinary water and in heavy water. However, for ordinary water, at least, this is not strictly the Fermi age, since the continuous slowing down model does not apply to media containing hydrogen. The result is actually the value of τ which approximately satisfies equation (3.93.1). It is roughly equivalent to $\overline{r_s^2}/6$ for thermal neutrons about a point source of fast neutrons, with which the age may be identified by equation (3.95.1). It will be seen later that this broader interpretation of neutron age is frequently useful.

* Attention may be called to the analogy between the neutron age and the diffusion length, as defined by equation (3.34.2).

SLOWING DOWN AND DIFFUSION PROPERTIES

Slowing Down and Migration Lengths

3.98. As indicated above, the Fermi age of neutrons is related to the mean square distance traveled while slowing down. The square root of the age is thus called the *slowing down length*. For thermal neutrons, of age τ_{th}, for example, $\sqrt{\tau_{th}}$ is a measure of the net vector (or crow-flight) distance traveled from their formation as fission neutrons to their attainment of thermal energy.

3.99. The sum of the square of the diffusion length of slow (thermal) neutrons, i.e., L^2, and of the age, τ, is called the *migration area*, M^2, i.e.,

$$M^2 \equiv L^2 + \tau, \tag{3.99.1}$$

and the square root is referred to as the migration length, M, i.e.,

$$M \equiv \sqrt{L^2 + \tau}. \tag{3.99.2}$$

Since, for thermal neutrons, L^2 is related to the mean square (net vector) distance the neutron travels from the point at which it becomes thermal to that at which it is captured, it is evident that the migration length for thermal neutrons is related to the total (net vector) distance covered between birth as a fission neutron and capture as a thermal neutron. The migration lengths for thermal neutrons in four common moderators, at ordinary temperatures, are recorded in Table 3.5. The diffusion and slowing down lengths are also included. For

TABLE 3.5. MIGRATION LENGTH OF THERMAL NEUTRONS

Moderator	*Diffusion Length* (cm)	*Slowing Down Length* (cm)	*Migration Length* (cm)
Water	2.88	5.74	6.43
Heavy water	100	10.9	101
Beryllium	23.6	9.90	25.8
Graphite	50.2	18.7	53.6

ordinary water and heavy water, M^2 is defined by

$$M^2 \equiv L^2 + \tfrac{1}{6}\overline{r_s^2}, \tag{3.99.3}$$

where $\overline{r_s^2}$ is the mean square slowing down distance from fission to thermal energies in these media. In fact, equation (3.99.3) may be regarded as the exact definition, applicable to all moderators, with equation (3.99.1) representing an alternative definition based on the continuous slowing down model. The migration length has a direct bearing on the size of a critical reactor, as mentioned in § 1.110.

Slowing Down and Diffusion Times

3.100. The average time spent by a neutron in slowing down to thermal energy and the diffusion time (or lifetime) of the thermal neutron before capture

may be readily calculated. Upon rearrangement, equation (3.88.1) takes the form

$$dt = -\frac{1}{v\xi\Sigma_s} \cdot \frac{dE}{E}, \tag{3.100.1}$$

where dt is the time required for the neutron energy to be decreased by an amount dE. The total time required for the neutron energy to be decreased from the fission energy E_0 to the thermal energy E_{th}, i.e., the mean chronological age, t, of the neutrons when they become thermal, is obtained upon integration of equation (3.100.1) over this range; thus,

$$t = \int_0^t dt = -\int_{E_0}^{E_{th}} \frac{1}{v\xi\Sigma_s} \cdot \frac{dE}{E}. \tag{3.100.2}$$

3.101. Since the energy of a neutron is kinetic, $E = \frac{1}{2}mv^2$, where m is the actual mass of a neutron; hence v may be replaced by $\sqrt{2E/m}$. Further, assuming an average value of Σ_s, represented by $\bar{\Sigma}_s$, equation (3.100.2) becomes

$$t = \frac{\sqrt{2m}}{\xi\bar{\Sigma}_s}\left(\frac{1}{\sqrt{E_{th}}} - \frac{1}{\sqrt{E_0}}\right). \tag{3.101.1}$$

In order to evaluate t in sec, $\bar{\Sigma}_s$ must be in cm^{-1}, as usual, m in grams (1.67×10^{-24} gram), and E_0 and E_{th} in ergs (1 Mev $\equiv 1.6 \times 10^{-6}$ erg). The values of $\bar{\Sigma}_s$ and the slowing down times from $E_0 = 2$ Mev to $E_{th} = 0.025$ ev, as calculated from equation (3.101.1), for four moderators, are given in Table 3.6.

TABLE 3.6. SLOWING DOWN AND DIFFUSION TIMES FOR THERMAL NEUTRONS

Moderator	$\bar{\Sigma}_s$ (cm^{-1})	*Slowing Down Time* (sec)	*Diffusion Time* (sec)
Water	0.90	10^{-5}	2.1×10^{-4}
Heavy water	0.43	2.9×10^{-5}	0.15
Beryllium	0.55	7.8×10^{-5}	4.3×10^{-3}
Graphite	0.30	1.9×10^{-4}	1.2×10^{-2}

3.102. The average diffusion time or *lifetime* of a thermal neutron is the mean time spent diffusing as a thermal neutron before capture by the moderator. In an infinite medium, the neutron lifetime (l_0) is equal to the absorption mean free path, i.e., the average (scalar) distance traveled before absorption, divided by the mean velocity v of the thermal neutrons; thus,

$$l_0 = \frac{\lambda_a}{v} = \frac{1}{v\Sigma_a}, \tag{3.102.1}$$

where Σ_a is the macroscopic absorption cross section for thermal neutrons. The mean velocity of these neutrons at ordinary temperatures is taken as 2.2×10^5 cm/sec, and the values of the lifetime (or diffusion time) are quoted in Table 3.6. It is seen that the average slowing down time in a moderator is usually

much less than the so-called lifetime of thermal neutrons. It should be noted that the results given are for infinite media, in which there is no leakage of neutrons from the system, and there is no absorption other than that by the moderator itself.

THE CRITICAL EQUATION

The Age-Diffusion Method

3.103. When a reactor is critical, a steady state is maintained without any extraneous source; consequently, in the steady-state equation (3.18.1), the source term S is now derived entirely from fission neutrons produced from the fuel material in the system. The number per cm³ per sec of such neutrons reaching any given energy is equal to the slowing down density at that energy. If there were no absorption during slowing down, this could be taken as equal to $q(\tau)$, which is the appropriate solution of the Fermi age equation.* As seen in § 3.92, for the case of weak absorption, the slowing down density is then given by $pq(\tau)$, where p is the resonance escape probability for neutrons of age τ. This quantity may, therefore, be substituted for S in equation (3.18.1). The treatment here will refer, in particular, to thermal neutrons, and so the steady-state diffusion equation for a specified point in the critical reactor is

$$D\nabla^2\phi(\mathbf{r}) - \Sigma_a\phi(\mathbf{r}) + pq(\tau_{\text{th}}, \mathbf{r}) = 0, \tag{3.103.1}$$

where ϕ is the thermal neutron flux and Σ_a is the macroscopic absorption cross section, at a point whose space coordinates are represented by the symbol $\mathbf{r}$ in general.

3.104. The slowing down density can be obtained by solving the age equation

$$\nabla^2 q(\tau, \mathbf{r}) = \frac{\partial q(\tau, \mathbf{r})}{\partial \tau} \tag{3.104.1}$$

with the appropriate conditions. One of these is determined by considering the value of the slowing down density of the fission (source) neutrons, i.e., $q(0, \mathbf{r})$, for which the age is zero.

3.105. In an infinite multiplying medium, consisting of a moderator and fuel material, for each thermal neutron absorbed in an infinite system there will be k thermal neutrons available in the next generation, k being the infinite multiplication factor of the medium (§ 1.44). The number of fission neutrons produced, for each thermal neutron absorbed, must then be k/p, where p is the resonance escape probability for thermal neutrons. The total number of thermal neutrons absorbed per cm³ per sec is $\Sigma_a\phi$, as given by the second term in equation (3.18.1); hence the total number of fission neutrons produced per cm³ per sec is $(k/p)\Sigma_a\phi$. This must represent the slowing down density for the fission neutrons, so that

* In the following treatment, it is more convenient to consider q as a function of age, rather than of energy.

$$q(0, \mathbf{r}) = \frac{k}{p}\Sigma_a\phi, \tag{3.105.1}$$

which provides a coupling condition between the thermal diffusion equation (3.103.1) and the age equation (3.104.1).

3.106. Since $q(\tau, \mathbf{r})$ is a function of the age and of the space coordinates, a solution of equation (3.104.1) may be sought by separating these variables, i.e., by writing

$$q(\tau, \mathbf{r}) = T(\tau)R(\mathbf{r}), \tag{3.106.1}$$

where $T(\tau)$ is a function of the age (or energy) only, and $R(\mathbf{r})$ is a function of the space coordinates only. Substitution of equation (3.106.1) into (3.104.1) then gives

$$\frac{\nabla^2 R(\mathbf{r})}{R(\mathbf{r})} = \frac{1}{T(\tau)} \cdot \frac{dT(\tau)}{d\tau}.$$

Since each side involves only one variable, it may be set equal to a constant; thus,

$$\frac{\nabla^2 R(\mathbf{r})}{R(\mathbf{r})} = -B^2$$

or

$$\nabla^2 R(\mathbf{r}) + B^2 R(\mathbf{r}) = 0 \tag{3.106.2}$$

and

$$\frac{1}{T(\tau)} \cdot \frac{dT(\tau)}{d\tau} = -B^2. \tag{3.106.3}$$

The solution of equation (3.106.2) is

$$T(\tau) = Ae^{-B^2\tau},$$

so that, from equation (3.106.1),

$$q(\tau, \mathbf{r}) = AR(\mathbf{r})e^{-B^2\tau}, \tag{3.106.4}$$

where B^2 must be a real positive quantity, in order to satisfy the physical requirement that the slowing down density cannot increase with neutron age.

3.107. When τ is zero, equation (3.106.4) becomes

$$q(0, \mathbf{r}) = AR(\mathbf{r}),$$

and, hence, from the coupling equation (3.105.1),

$$AR(\mathbf{r}) = \frac{k}{p}\Sigma_a\phi(\mathbf{r}). \tag{3.107.1}$$

Upon substituting this result back into equation (3.106.4), it follows that

$$q(\tau, \mathbf{r}) = \frac{k}{p}\Sigma_a\phi(\mathbf{r})e^{-B^2\tau}. \tag{3.107.2}$$

The critical thermal diffusion equation (3.103.1) can now be written as

$$D\nabla^2\phi(\mathbf{r}) - \Sigma_a\phi(\mathbf{r}) + k\Sigma_a\phi(\mathbf{r})e^{-B^2\tau_{th}} = 0$$

or, making use of the fact that L^2 is equal to D/Σ_a, by equation (3.33.2), it is readily found that

$$\nabla^2\phi(\mathbf{r}) + \frac{ke^{-B^2\tau_{th}} - 1}{L^2}\phi(\mathbf{r}) = 0. \tag{3.107.3}$$

3.108. According to equation (3.107.1), $\phi(\mathbf{r})$ is proportional to $R(\mathbf{r})$ and, hence, to $q(\mathbf{r}, \tau)$. If it is postulated that the extrapolation distance is independent of energy, so that the flux and the slowing down density become zero at the same boundary, it follows that $\phi(\mathbf{r})$ must satisfy the same equation as $R(\mathbf{r})$, namely, equation (3.106.2); consequently,

$$\nabla^2\phi(\mathbf{r}) + B^2\phi(\mathbf{r}) = 0, \tag{3.108.1}$$

where B^2, which is called the *buckling*,* is the same real positive quantity as above. Comparison of equations (3.107.3) and (3.108.1) then shows that

$$\frac{ke^{-B^2\tau_{th}} - 1}{L^2} = B^2,$$

or

$$\frac{ke^{-B^2\tau_{th}}}{1 + L^2B^2} = 1. \tag{3.108.2}$$

This is the *critical equation* for a bare, homogeneous reactor using what is called the age-diffusion method, i.e., diffusion theory applied to thermal neutrons with the spatial distribution of the slowing down density determined by the Fermi age.

3.109. In the foregoing treatment it has been tacitly assumed that the source neutrons, i.e., the fission neutrons, all have the same energy. However, equation (3.108.2) is applicable to the general case, provided τ_{th} is taken as the average value of the Fermi age of thermal neutrons properly weighted according to the fission spectrum (§ 2.166).

Buckling and Critical Size

3.110. The critical equation (3.108.2) relates the physical characteristics of the multiplying (reactor) system, as given by the quantities k, L^2, and τ, to the buckling, B^2. The latter, which is a geometrical quantity, dependent on the shape and size of the reactor, can be obtained by solving equation (3.108.1) for a specified shape, with the condition that the flux shall become zero at the extrapolated boundary. In this way it is possible to determine the critical size of a reactor of given shape from the physical properties of the materials present. The general treatment will be illustrated here by reference to a spherical reactor, but the results for other shapes will also be given.

* The name "buckling" arises from the fact that $-\nabla^2\phi(\mathbf{r})/\phi(\mathbf{r}) = B^2$ is a measure of the bending (or buckling) of the neutron flux at any point $\mathbf{r}$ in the critical reactor.

3.111. In order to solve the equation

$$\nabla^2\phi(\mathbf{r}) + B^2\phi(\mathbf{r}) = 0, \tag{3.111.1}$$

for a spherical reactor, it is convenient to choose the origin of the coordinates at the center of the sphere. Symmetry conditions then rule out any dependence of the neutron flux $\phi(\mathbf{r})$ on the angles θ and φ; hence, the Laplacian operator is derived from equation (3.16.3) by eliminating all terms involving these angles. Equation (3.111.1) for the present case thus takes the form

$$\frac{d^2\phi(r)}{dr^2} + \frac{2}{r}\cdot\frac{d\phi(r)}{dr} + B^2\phi(r) = 0. \tag{3.111.2}$$

Since B^2 is positive, the general solution of this differential equation is

$$\phi(r) = \frac{A}{r}\sin Br + \frac{C}{r}\cos Br,$$

where A and C are arbitrary constants. The fact that the flux is finite at the center of the sphere, i.e., when r is zero, eliminates the second term on the right, so that a permissible solution is

$$\phi(r) = \frac{A}{r}\sin Br. \tag{3.111.3}$$

3.112. If R is the radius of the spherical reactor, including the extrapolation distance, then the boundary condition referred to above requires that $\phi(r)$ shall be zero when $r = R$; hence, from equation (3.111.3),

$$\frac{A}{R}\sin BR = 0.$$

Since A and R are not zero, this means that $\sin BR = 0$, and consequently

$$BR = n\pi,$$

so that

$$B^2 = \left(\frac{n\pi}{R}\right)^2, \tag{3.112.1}$$

where n is an integer, i.e., 1, 2, 3, etc. Strictly speaking, $n = 0$ gives a permissible solution, but it is not included since it makes B^2 zero.

3.113. There are obviously an infinite number of values of B^2, corresponding to all integral values of n, which satisfy equation (3.111.1); these are referred to as the "eigenvalues" of this equation. However, the lowest eigenvalue, i.e., with $n = 1$, is the only one which is significant. It is the only value of B^2 which would make the neutron flux, as given by equation (3.111.3), positive for all values of r from zero to R. For $n > 1$, the solution oscillates, becoming negative for certain values of r. The buckling is thus taken as the lowest (nonzero) eigenvalue that results from solving equation (3.111.1), subject to the condition that the flux shall be zero at the extrapolated boundary of the system.

3.114. Taking $n = 1$, the buckling for the spherical reactor is given by

$$B^2 = \left(\frac{\pi}{R}\right)^2, \tag{3.114.1}$$

and the critical radius is then

$$R = \frac{\pi}{B}. \tag{3.114.2}$$

The flux distribution in the critical reactor is obtained by inserting the value for B from equation (3.114.1) into (3.111.3); thus,

$$\phi(r) = \frac{A}{r} \sin \frac{\pi r}{R}. \tag{3.114.3}$$

The arbitrary constant A in this expression is determined by the power level of the reactor, since this is proportional to the neutron flux (§ 2.163). As stated earlier, the power output of a reactor is, in principle, independent of its size, provided it is critical.

3.115. It will be seen from the foregoing treatment that, by prescribing the shape and size of a reactor, the value of the buckling can be determined, e.g., by equation (3.114.1) for a spherical reactor. If this result is now inserted into the critical equation (3.108.2), it is possible to determine the values of k, L^2, and τ, which depend upon the nature and proportions of fuel and moderator, that will make the prescribed reactor just critical. Alternatively, if the composition of the multiplying medium is specified, so that k, L^2, and τ are known, the buckling B^2 can be obtained from equation (3.108.2). The critical size for a given shape can then be determined, e.g., by equation (3.114.2) for a spherical reactor.

Reactors of Various Shapes

3.116. By solving equation (3.108.1) for reactors of various shapes, expressions for the buckling in terms of the appropriate spatial dimensions can be determined. The flux distribution in the critical reactor can then be derived in a manner similar to that described above for a sphere. The results are summarized in Table 3.7 for a sphere, a rectangular parallelepiped, and a finite cylinder, using the dimensions, which include the extrapolation distances, shown in Fig. 3.11.* The function J_0 is the Bessel function of the first kind of zero order. It will be noted that the buckling always has the dimensions of (length)$^{-2}$.

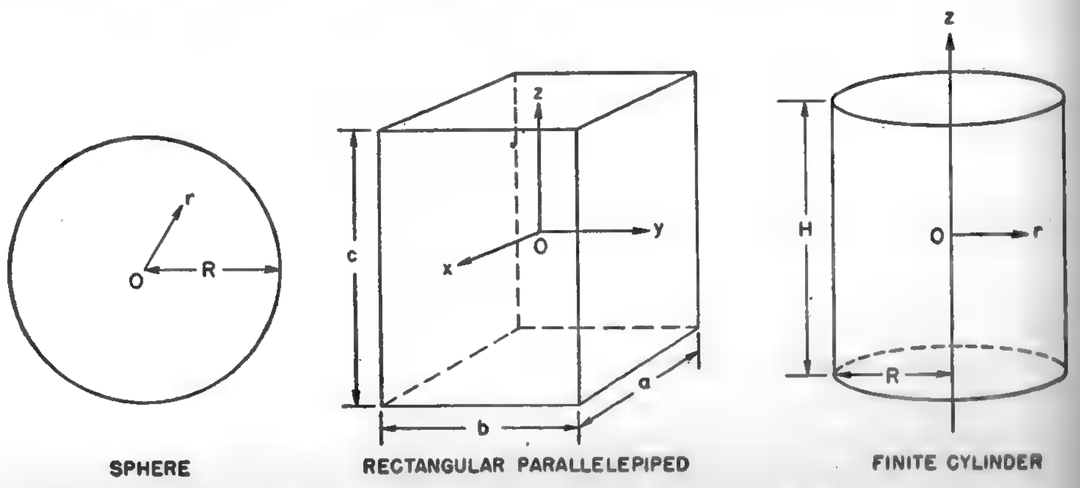


Fig. 3.11. Critical dimensions and coordinates for reactors of three geometries

TABLE 3.7. BUCKLING AND FLUX DISTRIBUTION IN BARE REACTORS

Geometry	Buckling	Critical Flux Distribution	Minimum Critical Volume
Sphere	$\left(\frac{\pi}{R}\right)^2$	$\frac{A}{r} \sin \frac{\pi r}{R}$	$\frac{130}{B^3}$
Rectangular parallelepiped	$\left(\frac{\pi}{a}\right)^2 + \left(\frac{\pi}{b}\right)^2 + \left(\frac{\pi}{c}\right)^2$	$A \cos \frac{\pi x}{a} \cos \frac{\pi y}{b} \cos \frac{\pi z}{c}$	$\frac{161}{B^3}$
Finite cylinder	$\left(\frac{2.405}{R}\right)^2 + \left(\frac{\pi}{H}\right)^2$	$AJ_0\left(\frac{2.405r}{R}\right) \cos \frac{\pi z}{H}$	$\frac{148}{B^3}$

3.117. It is seen that, in each case, the flux distribution is determined by one of three functions, namely, $(1/r) \sin (\pi r/R)$, $\cos (\pi x/a)$ or similar, and $J_0(2.405r/R)$. These all vary with the coordinates, i.e., with r, x (y, z, or H), and r, respectively, in a manner that is represented, to a very good approximation, by a cosine function (Fig. 3.12). Thus, the neutron flux distribution in a critical reactor may be assumed to have a cosine distribution about any of the

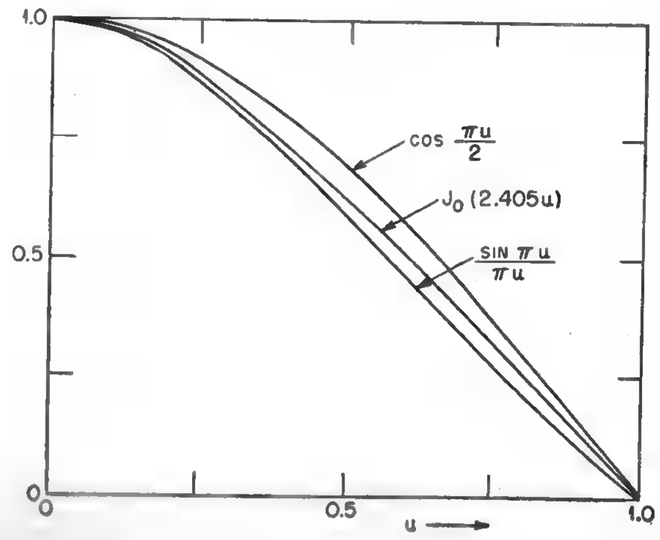


Fig. 3.12. Functions for determination of neutron flux distribution

* For details, see S. Glasstone and M. C. Edlund, *op. cit.*, Chapter VII.

axes in Fig. 3.11, with a relative value of unity at the axis and zero at the extrapolated boundary, in each case.

3.118. For a given value of the buckling, i.e., for a system of given composition, there will be a certain minimum volume for each kind of geometry which can be readily calculated from the appropriate buckling equations. The expressions for these minimum volumes are recorded in the last column of Table 3.7; in the case of the rectangular parallelepiped, the critical volume is least when the three sides are equal, i.e., for a cube. It is evident that, for a specified multiplying medium, the critical volume (and mass) of a spherical reactor will be less than for any of the other shapes. The reason is that the sphere has the smallest area-to-volume ratio. As stated in § 1.97, neutron production takes place throughout the volume of the reactor, but leakage occurs only at the surface. Hence, for a given composition, the reactor with the smallest ratio of surface to volume will have the smallest critical mass.

Nonleakage Probabilities

3.119. Before considering the applications of the critical equation (3.108.2), it is instructive to examine the significance of the terms involved. If a reactor were infinite in size, so that no neutrons were lost by leakage, the critical condition would be simply that the infinite multiplication factor k should be unity. Then for every neutron absorbed, another equivalent neutron would be produced. The use of the continuous slowing down model evidently introduces two factors, namely, $e^{-B^2\tau}$ and $1/(1 + L^2B^2)$, as is apparent from equation (3.108.2).* These take into account the fact that the reactor has finite dimensions, so that neutron leakage takes place.

3.120. According to equation (3.107.2), the slowing down density of source (fission) neutrons is $(k/p)\Sigma_a\phi$, since τ is zero, and the slowing down density of thermal neutrons, disregarding absorption, is $(k/p)\Sigma_a\phi e^{-B^2\tau}$. If there had been no leakage while slowing down, these two quantities would have been equal. Hence $e^{-B^2\tau}$ is the fraction of the source neutrons that remain in the reactor and do not escape. In other words, $e^{-B^2\tau}$ represents the *nonleakage probability* (cf. § 1.95) of the neutrons during the course of slowing down from source to age τ.

3.121. The rate at which neutrons leak away from a specified point in the reactor is given by diffusion theory as $-D\nabla^2\phi$, and by equation (3.108.1) this may be taken as equal to $DB^2\phi$ neutrons/(cm³)(sec). The rate of absorption is $\Sigma_a\phi$ thermal neutrons/(cm³)(sec), and so the ratio of thermal leakage to thermal absorption is given by

$$\frac{\text{Thermal leakage}}{\text{Thermal absorption}} = \frac{DB^2}{\Sigma_a} = L^2B^2,$$

since D/Σ_a is equal to L^2, the square of the thermal diffusion length. By adding

* Since it is understood that τ refers to the age of the thermal neutrons, the subscript will be omitted for simplicity.

unity to each side and inverting, it is seen that

$$\frac{\text{Thermal absorption}}{\text{Thermal absorption + thermal leakage}} = \frac{1}{1 + L^2B^2}. \qquad (3.121.1)$$

Since the denominator on the left side represents the total number of neutrons thermalized, it follows that $1/(1 + L^2B^2)$ is the fraction of neutrons thermalized in the reactor that are absorbed, while the remainder leak out of the reactor. Consequently $1/(1 + L^2B^2)$ is the nonleakage probability for thermal neutrons.

3.122. It is of interest to consider the physical significance of the quantities appearing in the nonleakage probability factor. In view of the inverse relationship between B^2 and the dimensions of the reactor (see Table 3.7), B^2 will be large when the reactor is small, and vice versa. A large value of B^2 will mean that both $e^{-B^2\tau}$ and $1/(1 + L^2B^2)$ are relatively small. Thus, as is to be expected, the neutron leakage, both during slowing down and while thermal, will be large for a small reactor. Next, consider the age τ; this is related to the average (net vector) distance traveled by a neutron while slowing down. It is to be anticipated, therefore, that for a reactor of given size, a large τ will mean a small nonleakage probability; this is in harmony with the results obtained above, for $e^{-B^2\tau}$ is then small. Similar considerations apply to the diffusion length, L, which is a measure of the average (net vector) distance traveled by the thermal neutron before capture. A large value of L should mean a large probability of leakage from the reactor, and this is in agreement with the thermal nonleakage probability represented by $1/(1 + L^2B^2)$.

3.123. Since B^2 is inversely related to the reactor size, it is evident that for a *large reactor* the exponential $e^{-B^2\tau_{th}}$ in the numerator of the age-diffusion critical equation (3.108.2) can be expanded in series form and all terms beyond the second neglected, so that

$$e^{-B^2\tau} \approx 1 - B^2\tau \approx (1 + B^2\tau)^{-1},$$

it being understood that τ refers to thermal neutrons. In these circumstances the critical equation becomes

$$\frac{k}{(1 + \tau B^2)(1 + L^2B^2)} \approx 1. \qquad (3.123.1)$$

Neglecting terms in B^4, as being very small, the critical equation for a large reactor may be written as

$$\frac{k}{1 + B^2(L^2 + \tau)} \approx 1. \qquad (3.123.2)$$

As seen in § 3.99, the sum $L^2 + \tau$, or $L^2 + \frac{1}{6}\overline{r_s^2}$, in general, is defined as the migration area M^2 for thermal neutrons, so that critical equation (3.123.2) for a large reactor is

$$\frac{k}{1 + M^2B^2} \approx 1. \qquad (3.123.3)$$

3.124. According to this result, the total nonleakage probability is given by $1/(1 + M^2B^2)$; the greater the value of M^2 the smaller is the nonleakage probability, i.e., the greater the neutron loss by leakage, for a reactor of specified size. Since the migration length includes both slowing down and thermal diffusion lengths, this result is to be expected. It is of interest to note that equation (3.123.3) can be written in the form $k - 1 \approx M^2B^2$, and since B^2 is equal to $(b/R)^2$, where R is the radius of a sphere or the edge length of a cube and b depends on the shape of the reactor, this result is equivalent to equation (1.111.2), derived in a general manner in Chapter I.

Example A cubical thermal reactor, in which uranium-235 is the fuel and beryllium the moderator, has an edge length of 100 cm. What proportion of the neutrons produced in fission are lost by escape? (The Fermi age of thermal neutrons may be taken to be that in beryllium, i.e., 98 cm² at 20°C, but L^2 is only about 70 cm²; see § 3.147.)

The nonleakage probability while slowing down is $e^{-B^2\tau}$, and so the corresponding leakage probability is $1 - e^{-B^2\tau}$; this is the fraction of neutrons that escape while slowing down. Since a cube is a rectangular parallelepiped with equal edges, it follows from Table 3.7 that $B^2 = 3(\pi/a)^2$, where a is the edge length. In the present case a is 100 cm and so B^2 is 2.9×10^{-3} cm^{-2}. Hence,

$$1 - e^{-B^2\tau} = 1 - e^{-(2.9\times10^{-3})(98)} = 0.25,$$

so that 0.25 of the fission neutrons leak out while slowing down and the remaining 0.75 is thermalized.

The nonleakage probability of thermal neutrons is $1/(1 + L^2B^2)$, and so the leakage probability is $L^2B^2/(1 + L^2B^2)$. The value of L^2 in the reactor is given as 70 cm²; L^2B^2 is then 0.20, and the fraction of thermal neutrons lost by leakage is 0.20/1.20, i.e., 0.17. Since only 0.75 of the fission neutrons are thermalized, the fraction of fission neutrons escaping as thermal neutrons is (0.17)(0.75) = 0.13. The total loss of fission neutrons by leakage is thus 0.25 + 0.13 = 0.38, or 38 per cent.

If the reactor had been treated as large, so that the total nonleakage probability would be represented by $1/(1 + M^2B^2)$ and the fraction of neutrons lost by leakage by $M^2B^2/(1 + M^2B^2)$, the latter would be 0.33, or 33 per cent. (Note that $M^2 = L^2 + \tau$ is here 70 + 98 = 168 cm².)

The Effective Multiplication Factor

3.125. The product of the two terms $e^{-B^2\tau}$ and $1/(1 + L^2B^2)$ is equal to the total nonleakage probability of neutrons in a reactor of finite size, as derived from age-diffusion theory. This is the quantity represented by P in § 1.95; hence, $ke^{-B^2\tau}/(1 + L^2B^2)$ must, by definition, be equal to the effective multiplication factor, i.e.,

$$\frac{ke^{-B^2\tau}}{1 + L^2B^2} \equiv k_{\text{eff}}. \tag{3.125.1}$$

For a critical reactor, of finite size, k_{eff} is equal to unity, and equation (3.125.1) then becomes identical with the critical equation (3.108.2).

3.126. If the reactor is larger than the critical size for the given materials and composition, i.e., the reactor is supercritical, the value of B^2, as determined from the geometry of the system, will be less than that for a critical reactor, because

B^2 is related inversely to the reactor dimensions, as seen above. The left side of equation (3.125.1) will then exceed unity, so that $k_{\text{eff}} > 1$, as required for a supercritical system (§ 1.94). In this case, the two nonleakage probabilities, $e^{-B^2\tau}$ and $1/(1 + L^2B^2)$, will both be greater than required to make the reactor critical. If the reactor is subcritical, so that B^2 is greater than the critical value, k_{eff}, as given by equation (3.125.1), will be less than unity, because the nonleakage probabilities are then smaller than necessary for the given system to become critical.

Neutron Lifetime in Finite Reactors

3.127. It was found in § 3.102 that the mean lifetime of a thermal neutron in an infinite medium, where the loss of neutrons is due entirely to absorption, is given by $l_0 = 1/\Sigma_a v$. However, in a system of finite size, the effective lifetime is decreased because neutrons are also lost by leakage. According to equation (3.121.1), the ratio of neutrons absorbed to the sum of those absorbed and leaking away is $1/(1 + L^2B^2)$; consequently the average thermal neutron lifetime, l, in a finite reactor is

$$l = \frac{l_0}{1 + L^2B^2}. \tag{3.127.1}$$

3.128. For a large thermal reactor, using natural uranium as fuel, the quantity $1 + L^2B^2$ is not much greater than unity, and so the finite lifetime is not very different from that in an infinite system of the same composition. If fuel enriched in uranium-235 is used, as it is in several modern reactors, B^2 is much larger than for a natural-uranium reactor. The mean lifetime of thermal neutrons in the finite reactor may then be appreciably smaller than in the infinite system. The actual values range from about 10^{-3} sec for natural-uranium reactors to roughly 10^{-4} sec for some using enriched fuel (see § 3.164).

The Multiplication Factor for Thermal Reactors

3.129. The infinite multiplication factor k of the materials is probably the most important quantity in relation to reactor criticality. Unless k for the given multiplying medium exceeds unity, a critical system of finite size is impossible. If k is only slightly greater than unity, then the critical reactor will have to be large, i.e., B^2 must be small, so as to keep the neutron leakage to a minimum. On the other hand, if k is relatively large, then a small critical reactor will be possible, and B^2 can be large. The nonleakage probabilities can then be relatively small, i.e., the fractional loss of neutrons by leakage can be considerable, and yet the product of k and the nonleakage probability factor can still be equal to (or exceed) unity.

3.130. The multiplication factor for an infinite system, i.e., with no leakage, may conveniently be divided into four parts, in the following manner. As stated in § 1.104, η is the average number of fast (fission) neutrons emitted as a result of the capture of a thermal neutron in fuel, i.e., in both uranium-235 and

uranium-238 in the case of a mixture of these two isotopes. Since not all neutrons absorbed in fuel cause fission, the average number of fast (fission) neutrons released per thermal-neutron fission is ν, which, by equation (1.104.1), is related to η by

$$\eta = \nu \times \frac{\text{Neutrons causing fission}}{\text{Total neutrons absorbed in fuel}}$$
$$= \nu \times \frac{\Sigma_f}{\Sigma_u},$$

where Σ_f is the thermal-neutron fission cross section of the fissionable material, e.g., uranium-235, and Σ_u is the total cross section for absorption of thermal neutrons, by fission and nonfission processes, in the fuel material, e.g., natural uranium. Some values for η were recorded in Table 1.7. For thermal-neutron fission in natural uranium, for example, η is 1.3, so that for every neutron absorbed in this fuel, 1.3 fast (fission) neutrons are produced.

3.131. Suppose that at a given instant, representing the initiation of a generation, there are available n thermal neutrons which are absorbed in fuel; then, as a result, $n\eta$ fast neutrons will be produced by fission. Before these fast neutrons have slowed down appreciably some will be captured by, and cause fission of, uranium-235 and uranium-238 nuclei. At neutron energies greater than about 1 Mev, most of the fast-neutron fissions in natural uranium will be of uranium-238 nuclei because of their greater proportion. Since more than one neutron is produced, on the average, in each fast-neutron fission, there will be an increase in the number of neutrons available. Allowance for this effect may be made by introducing the *fast-fission factor*, denoted by ϵ and defined as the ratio of the total number of fast neutrons produced by fissions due to neutrons of all energies (fast and thermal) to the number resulting from thermal-neutron fissions. Consequently the capture of n thermal neutrons in fuel will cause $n\eta\epsilon$ fast neutrons to be liberated.*

3.132. During slowing down some of the neutrons are captured in nonfission processes, and the fraction escaping capture is p, the resonance escape probability. Hence, the number of neutrons which become thermalized is $n\eta\epsilon p$. According to equation (3.79.2), the resonance escape probability depends on the macroscopic absorption and scattering cross sections of the fuel-moderator system. It is thus determined both by the nature of the materials and their proportions. Some values of p for mixtures of graphite and natural uranium will be given later.

3.133. When the neutrons have reached thermal energies, they will diffuse for some time in the infinite system until they are ultimately absorbed by fuel, by moderator, or by such impurities (poisons) as may be present. Of the thermal neutrons, therefore, a fraction f, called the *thermal utilization*, will be absorbed

* The value of ϵ in reactors using natural uranium as fuel, with either graphite or heavy water as moderator, is about 1.03 (see § 3.176). In homogeneous (uniform) systems it is approximately unity.

in fuel material; the value of f is represented by

$$f = \frac{\text{Thermal neutrons absorbed in fuel}}{\text{Total thermal neutrons absorbed}}$$
$$= \frac{V_u\Sigma_u\phi_u}{V_u\Sigma_u\phi_u + V_m\Sigma_m\phi_m + V_i\Sigma_i\phi_i}, \qquad (3.133.1)$$

where the subscripts u, m, and i indicate fuel (uranium), moderator, and impurities, respectively. The various Σ's refer to absorption cross sections for thermal neutrons, and the V's to the respective volumes occupied by the indicated substances. Like the resonance escape probability, the values of f depend on the fuel and moderator and on their relative amounts.

3.134. Since all the $n\eta\epsilon p$ neutrons reaching thermal energies in the infinite system are absorbed, the number absorbed in fuel will be $n\eta\epsilon pf$. The infinite multiplication factor k is defined as the ratio of the average number of thermal neutrons produced (and hence absorbed) in one generation to the number of thermal neutrons produced (or absorbed) in the preceding generation, in an infinite medium (§ 1.95); consequently

$$k = \frac{n\eta\epsilon pf}{n} = \eta\epsilon pf. \qquad (3.134.1)$$

This is sometimes referred to as the *four-factor formula*. The division of the infinite multiplication factor into four parts simplifies the procedure for evaluating this important property of a multiplying system.

HOMOGENEOUS SYSTEMS

Resonance Escape Probability and Thermal Utilization

3.135. In the equation (3.79.2), which will be used here to express the resonance escape probability in a homogeneous system, Σ_a may be assumed to refer to the fuel only, since neutron absorption in the moderator is, in general, relatively small. Hence Σ_a may be replaced by $N_u\sigma_{au}$, where N_u is the number of atoms (or nuclei) per cm³ of the fuel, and σ_{au} is the absorption cross section for neutrons of energy E. It is then possible to write

$$\frac{\Sigma_a}{\Sigma_a + \Sigma_s} = \frac{N_u\sigma_{au}}{\Sigma_s} \cdot \frac{\Sigma_s}{\Sigma_a + \Sigma_s}.$$

Since, in the resonance region, the scattering cross section may be taken to be independent of neutron energy, equation (3.79.2) becomes

$$p(E) = \exp\left[-\frac{N_u}{\xi\Sigma_s}\int_E^{E_0}\left(\frac{\Sigma_s}{\Sigma_a + \Sigma_s}\,\sigma_{au}\right)\frac{dE}{E}\right]. \qquad (3.135.1)$$

The integral in this equation is called the *effective resonance integral*,* and it can be represented by

* The actual or true resonance integral for any substance is the integral of $\sigma_a\,dE/E$ over the resonance energy range.

$$\text{Effective resonance integral} = \int (\sigma_{au})_{\text{eff}} \frac{dE}{E}, \tag{3.135.2}$$

where

$$(\sigma_{au})_{\text{eff}} = \frac{\Sigma_s}{\Sigma_a + \Sigma_s} \sigma_{au},$$

so that

$$p(E) = \exp\left[-\frac{N_u}{\xi \Sigma_s} \int (\sigma_{au})_{\text{eff}} \frac{dE}{E} \right], \tag{3.135.3}$$

the integration being carried over the resonance energy range.

3.136. It is seen from equation (3.135.3) that the resonance escape probability depends upon the effective resonance integral. The latter can be determined experimentally for a given mixture of fuel and moderator. For mixtures of natural uranium with several different moderators, it has been found that the effective resonance integral is expressed to a good approximation by the formula

$$\text{Effective resonance integral} = 3.9 \left(\frac{\Sigma_s}{N_u} \right)^{0.415} \text{barns}, \tag{3.136.1}$$

the limiting value for great dilutions, as N_u approaches zero, being 240 barns. With the effective resonance integral known, the resonance escape probability can be calculated by means of equation (3.135.3). As the proportion of fuel (natural uranium) in the system increases, it is seen that the resonance escape probability decreases, as is to be expected (see Table 3.8, below).

Example Determine the resonance escape probability in a uniform mixture of graphite and natural uranium metal in the atomic ratio (N_m/N_u) of 400. (The scattering cross section for resonance neutrons may be taken as 4.8 barns.)

By combining equations (3.135.3) and (3.136.1) it is seen that for natural uranium as fuel,

$$p(E) = \exp\left[-\frac{3.9}{\xi} \left(\frac{N_u}{\Sigma_s} \right)^{0.585} \right].$$

Because of the large proportion of graphite, ξ may be taken as the value for carbon, i.e., 0.158, and Σ_s may be set equal to $N_m \sigma_{sm}$, where σ_{sm} is 4.8 barns; thus

$$\begin{aligned} p(E) &= \exp\left[-\frac{3.9}{\xi} \left(\frac{N_u}{N_m \sigma_{sm}} \right)^{0.585} \right] \\ &= \exp\left[-\frac{3.9}{0.158} \left(\frac{1/400}{4.8} \right)^{0.585} \right] \\ &= e^{-0.296} = 0.744. \end{aligned}$$

The required resonance escape probability is thus 0.74_4, since it is doubtful if the third figure has any significance.

3.137. For a uniform mixture of fuel and moderator the neutron flux is the same throughout and all the constituents occupy the same volume, so that equation (3.133.1) for the thermal utilization reduces to

$$f = \frac{\Sigma_u}{\Sigma_u + \Sigma_m + \Sigma_i},$$

where the Σ's are absorption cross sections. In the absence of impurities (poisons) this result may be written as

$$f = \frac{\Sigma_{au}}{\Sigma_{au} + \Sigma_{am}} = \frac{N_u \sigma_{au}}{N_u \sigma_{au} + N_m \sigma_{am}}, \tag{3.137.1}$$

where, as above, the N's are the respective numbers of nuclei per cm³. The σ_a's are now the microscopic absorption cross sections for *thermal* neutrons. In contrast to the variation of the resonance escape probability with composition, it is seen from equation (3.137.1) that the thermal utilization increases with increasing proportion of fuel. It would be unity for pure fuel, in the absence of moderator, i.e., when N_m is zero.

Example Determine the thermal utilization for the uniform mixture of natural uranium and graphite considered in the preceding example, for which N_m/N_u is 400. (The microscopic absorption cross sections of natural uranium and graphite for thermal neutrons are 7.42 barns and 0.0045 barn, respectively.)

Equation (3.137.1) may also be written as

$$f = \frac{\sigma_{au}}{\sigma_{au} + \sigma_{am}(N_m/N_u)}$$

and since N_m/N_u is 400, it follows that

$$f = \frac{7.42}{7.42 + (0.0045)(400)} = 0.804.$$

The required value of the thermal utilization is thus 0.80_4, the third figure being doubtful.

3.138. Some values for the resonance escape probability and the thermal utilization for various uniform (homogeneous) mixtures of natural uranium metal and graphite are given in Table 3.8. As the proportion of fuel decreases, i.e., as N_m/N_u increases, p increases but f decreases. The product pf passes through a maximum of about 0.60, when there is roughly one atom of uranium to 400 atoms of graphite in the mixture.

TABLE 3.8. RESONANCE ESCAPE PROBABILITY AND THERMAL UTILIZATION IN UNIFORM MIXTURES OF NATURAL URANIUM AND GRAPHITE

N_m/N_u	p	f	pf
200	0.64	0.89	0.57
300	0.70	0.84	0.59
400	0.74	0.80	0.60
500	0.77	0.76	0.59

3.139. It was seen above that, for natural uranium as fuel, the value of η is 1.3, and since ϵ is approximately unity for a homogeneous mixture, the maximum

value of the infinite multiplication factor, i.e., $\eta\epsilon pf$, for a homogeneous mixture of natural uranium and graphite is 1.3×0.60, i.e., about 0.80. Since this is less than unity it is obviously impossible to establish a self-sustaining chain reaction in this system. In other words, no matter how large a reactor were constructed, it would never go critical.*

3.140. There are two ways in which the problem of obtaining a critical system with uranium as fuel has been solved. One is to use enriched fuel, i.e., fuel containing an appreciably greater proportion of the fissionable uranium-235 than does natural uranium. The other is to take advantage of the fact that the resonance escape probability, for a given ratio of fuel to moderator, can be increased by using a heterogeneous lattice system consisting of lumps (or rods) of natural uranium arranged in a matrix of graphite.

Highly Enriched Fuel Systems†

3.141. The use of enriched fuel results in a decrease in the proportion of neutrons absorbed in nonfission reactions in uranium-238, both as thermal neutrons and while slowing down. There is thus an increase in the infinite multiplication factor due to increases in η and p, in particular, and also, frequently, in the thermal utilization. The effect of enriched fuel on the fast-fission factor is small. Since, in any event, ϵ is close to unity in a homogeneous system, the change may be neglected.

3.142. If the fuel consists of pure fissionable material, e.g., uranium-235, then η is given by equation (1.105.2) as

$$\eta = \frac{\nu}{1 + \alpha},$$

with $\alpha = \sigma_c/\sigma_f$, where σ_c is the nonfission capture cross section for thermal neutrons, and σ_f is the cross section for fission. It is seen from Table 2.10 that, for uranium-235, σ_c is 101 barns and σ_f is 549 barns, and since ν is 2.5, it follows that

$$\eta = \frac{2.5}{1 + \dfrac{101}{549}} = 2.1,$$

which is the result given in Table 1.7. This represents a considerable increase over the value for natural uranium.

3.143. A reactor employing enriched material can be made critical with a very low concentration of fuel. Because of the large ratio of moderator to fuel, the fraction of neutrons captured in fuel while slowing down is small, since the probability of scattering is large compared to absorption. The resonance escape probability is then not much less than unity, and as η is slightly greater than unity, it is a good approximation to write

$$k = \eta f. \tag{3.143.1}$$

3.144. Since η is 2.1, the thermal utilization could be as low as 0.5, i.e., $k = 1.05$, and the system still be capable of becoming critical. For a homogeneous mixture of fuel and moderator, in the absence of impurities, f is given by equation (3.137.1), and the system would be capable of maintaining a chain reaction even when $N_u\sigma_{au} = N_m\sigma_{am}$, so that f is 0.5. For uranium-235, σ_{au} is $549 + 101 = 650$ barns, and if graphite is the moderator, σ_{am} is 0.0045 barn. Hence, in these circumstances, N_m/N_u would be 1.44×10^5. This means that a homogeneous mixture consisting of 1 atom of uranium-235 to roughly 144,000 atoms of graphite could be used in a thermal reactor. Since k is only 1.05, the critical reactor would, of course, be large. If the ratio of uranium-235 to moderator is increased, the thermal utilization is increased, and so also is the infinite multiplication factor. Criticality could then be attained with a reactor of smaller size.

Calculation of Critical Size

3.145. The first step in the calculation of the critical size of a reactor is to determine the corresponding buckling. For a homogeneous system, a first approximation may be derived relatively simply by neglecting the neutron absorption due to impurities, e.g., structure, coolant, poisons, etc. If the fuel is highly enriched, it is sufficiently accurate for the present purpose to assume that $k = \eta f$, as in equation (3.143.1). Further, if the ratio of the macroscopic absorption cross sections of fuel to moderator is represented by z, i.e.,

$$z \equiv \frac{\Sigma_{au}}{\Sigma_{am}} = \frac{N_u\sigma_{au}}{N_m\sigma_{am}}, \tag{3.145.1}$$

it follows from equation (3.137.1) that

$$f = \frac{z}{z + 1}, \tag{3.145.2}$$

and, consequently,

$$k = \frac{\eta z}{z + 1}. \tag{3.145.3}$$

3.146. As seen above, an enriched system can be critical even when the proportion of fuel is very small. Hence, in the uniform fuel-moderator, the diffusion coefficient and age of thermal neutrons will be essentially the same as in pure moderator. By equation (3.34.1), $L^2 = D/\Sigma_a$, and since D may be taken as the diffusion coefficient of the pure moderator, i.e., D_m, it follows that

$$L^2 = \frac{D_m}{\Sigma_{au} + \Sigma_{am}}.$$

* By using a better moderator than graphite, e.g., heavy water, the value of k could be increased to the extent that a uniform mixture with natural uranium can become critical. The large size of the reactor coupled with the high cost of heavy water makes such a system rather impractical.

† For a treatment of slightly enriched fuel systems, see § 3.184.

3.147. If this result is combined with equation (3.137.1), which defines f, it is found that

$$L^2 = \frac{D_m}{\Sigma_{am}}(1 - f) = L_m^2(1 - f) \tag{3.147.1}$$

or, in view of equation (3.145.2),

$$L^2 = \frac{L_m^2}{z + 1}, \tag{3.147.2}$$

where L_m^2 refers to the pure moderator.

3.148. Upon introducing the values of k and L^2 given by equations (3.145.3) and (3.147.2) into the critical equation (3.108.2), it is seen that

$$\frac{\eta z e^{-B^2\tau_m}}{z + 1 + L_m^2B^2} = 1, \tag{3.148.1}$$

where τ_m and L_m^2 refer to the pure moderator. For a fuel-moderator mixture of specified composition, η, z, L_m^2, and τ_m may be regarded as known, and so the buckling, B^2, can be evaluated by solving the transcendental equation (3.148.1).

3.149. A convenient method of solution is to write the equation in the form

$$e^{-B^2\tau_m} = \frac{1}{\eta z}(z + 1 + L_m^2B^2), \tag{3.149.1}$$

and let

$$A \equiv \frac{z + 1}{\eta z} \qquad C \equiv \frac{L_m^2}{\eta z \tau_m} \qquad x \equiv B^2\tau_m,$$

where A and C are constants, and x is a variable. The critical equation (3.149.1) then becomes

$$e^{-x} = A + Cx. \tag{3.149.2}$$

Since x is not large, e^{-x} may be taken as approximately equal to $1 - x$; then, A and C being known, a preliminary value of x is obtained by writing equation (3.149.2) as $x = (1 - A)(C + 1)$. With this x, the two sides of equation (3.149.2) are evaluated. In general, they will not be equal, and so another guess is made until, by a process of iteration, the value of x is found which satisfies equation (3.149.2). Since τ_m is known, this gives B^2, as required.*

3.150. In order to determine the dimensions of the critical reactor, the value of B^2 just obtained is set equal to the appropriate expression in Table 3.7 for the required shape. The critical volume and dimensions for this shape can then be readily calculated.

Example Calculate the critical volume (at ordinary temperature) of a spherical reactor containing beryllium and uranium-235 in the atomic ratio of 10^4 to 1.

The basic data for thermal neutrons at ordinary temperature are as follows:

* For a graphical method of solving the critical equation, see F. T. Miles and H. Soodak, *Nucleonics*, 11, No. 1, 66 (1953).

For beryllium:
$L_m = 23.6$ cm (Table 3.1)
$\tau_m = 98$ cm² (Table 3.4)
$\sigma_{am} = 0.009$ barn (Table 2.7).

For uranium-235:
$\sigma_{au} = 650$ barns (Table 2.10)
$\eta = 2.1$ (Table 1.7).

In addition, N_m/N_u is given as 10^4. Consequently,

$$\alpha = \frac{N_u\sigma_{au}}{N_m\sigma_{am}} = \frac{650}{(10^4)(0.009)} = 7.2$$

$$A = \frac{z + 1}{\eta z} = \frac{(7.2 + 1)}{(2.1)(7.2)} = 0.54$$

$$C = \frac{L_m^2}{\eta z \tau_m} = \frac{(23.6)^2}{(2.1)(7.2)(98)} = 0.37.$$

The preliminary value of $x = (1 - A)(C + 1)$ is thus 0.63, but insertion into equation (3.149.2) shows the right side to be too large, so that this value of x is also too large. By a process of iteration $x = B^2\tau_m$ is found to be 0.37, so that

$$B^2 = \frac{0.37}{98} = 3.8 \times 10^{-3}\ \text{cm}^{-2}.$$

From Table 3.7, the critical volume of a sphere is related to the buckling by

$$V = \frac{130}{B^3} = \frac{130}{(3.8 \times 10^{-3})^{\frac{3}{2}}} = 5.4 \times 10^5\ \text{cm}^3,$$

which is roughly 20 ft³. The radius is readily calculated to be about 50 cm or just less than 20 in., including the extrapolation distance.

3.151. A simpler type of problem which can be solved with the same equations is to determine the composition of a given fuel-moderator system which will be critical for a specified size and shape. Introducing the definition of z, as given in § 3.145, equation (3.148.1) can be rearranged to

$$\frac{N_m}{N_u} = \frac{\eta e^{-B^2\tau_m} - 1}{1 + L_m^2B^2} \cdot \frac{\sigma_{au}}{\sigma_{am}}. \tag{3.151.1}$$

Since B^2 can be determined from the specified geometry of the reactor, and the physical properties σ_{au}, σ_{am}, L_m^2, η, and τ_m may be regarded as known for the reactor materials, the composition of the system, as expressed by N_m/N_u, can be readily evaluated.

3.152. As an aid to calculations and in order to give some indication of the magnitudes involved, N_m/N_u has been determined as a function of B^2, by means of equation (3.151.1), for pure uranium-235 as fuel and either heavy water, beryllium, or carbon (graphite) as moderator. The results are plotted in Fig. 3.13.* (A curve for ordinary water as moderator is also given, but this will be discussed later, since it is derived in a different manner.) In the case of heavy water as moderator, N_m refers to the number of molecules of this substance; for

* Based on calculations made by M. C. Edlund.

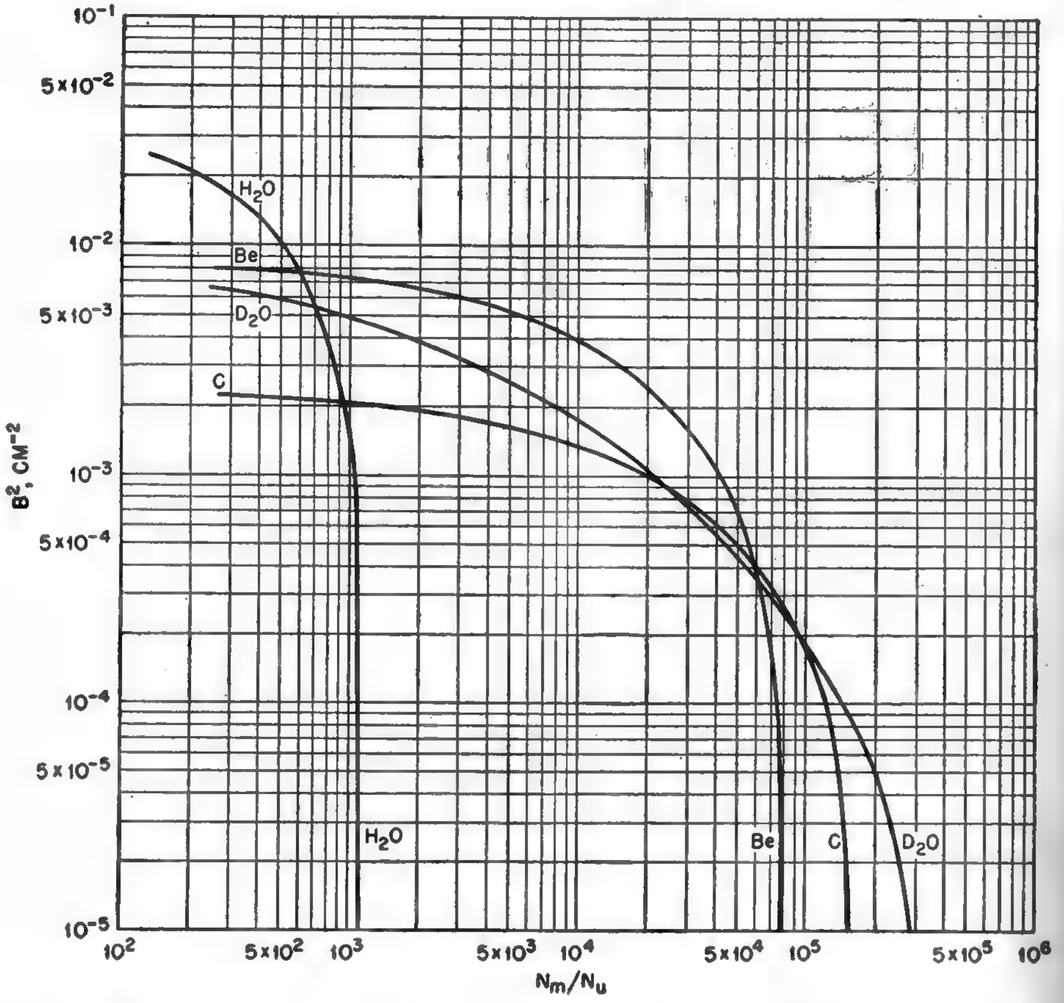


FIG. 3.13. Relationship between buckling and composition of thermal reactors with pure uranium-235 as fuel

the other moderators it represents the number of atoms. If the composition of the fuel-moderator system, i.e., N_m/N_u, is specified, then B^2 can be obtained from the figure; the critical dimensions for a given shape can then be calculated. On the other hand, if the geometry is specified, B^2 is known and the composition required to make the system critical can be read from the curves.

MINIMUM CRITICAL MASS

3.153. It can be seen from Fig. 3.13 that as the ratio of moderator to fuel is increased, the dimensions of the critical reactor must increase. It is of interest to note, however, that by varying the composition of the mixture, i.e., by changing the moderator-to-fuel ratio, it is possible to obtain a system which will become critical with a minimum mass of fuel. It will be shown, in the first

place, that there are two conditions under which the mass of fuel becomes infinite.

3.154. One occurs when k is unity, so that the critical reactor is infinitely large, and B^2 is zero. In these circumstances, equation (3.145.3) reduces to

$$\frac{\eta z}{z+1} = 1,$$

so that

$$z = \frac{1}{\eta - 1}.$$

The same result, of course, can be obtained by writing $k = \eta f = 1$, where f is given by equation (3.145.2). In view of the definition of z by equation (3.145.1), it follows that the critical reactor becomes infinitely large when

$$\frac{N_m}{N_u} = \frac{\sigma_{au}(\eta - 1)}{\sigma_{am}}. \tag{3.154.1}$$

If N_m/N_u is greater than this, the system cannot maintain a chain reaction because of excessive neutron absorption in the moderator. For a homogeneous mixture of uranium-235 and graphite, for example, σ_{au} is 650 barns, σ_{am} is 0.0045 barn, and $\eta - 1$ is 1.1; the limiting value of the moderator-to-fuel atomic ratio is thus 1.37×10^5 to 1.

3.155. The existence of this limiting ratio of moderator to fuel is also apparent from Fig. 3.13. It is seen that the curves fall off steeply to the right and tend toward an asymptotic value of N_m/N_u as B^2 decreases and the reactor dimensions increase. If the proportion of fuel is less than the limiting value, the reactor cannot be made critical, no matter how large it is and what mass of fuel it contains. It may be noted, incidentally, that according to equation (3.154.1) the asymptotic values of N_m/N_u are inversely related to the microscopic absorption cross sections of the moderators; the latter are in the order: water > beryllium > graphite > heavy water, in agreement with the positions of the curves in Fig. 3.13.

3.156. The other condition for infinite mass of fuel is that N_m/N_u should be zero, as implied by the flattening of the curves at the left of Fig. 3.13. It is seen from equation (3.151.1) that the ratio of N_m to N_u will become zero when $\eta e^{-B^2\tau_m} = 1$. The corresponding asymptotic value of B^2 is then inversely proportional to τ_m, the age of thermal neutrons in the moderator. The latter follow the order: graphite > heavy water > beryllium > water, so that, in accordance with Fig. 3.13, the asymptotic values of the buckling are in the reverse order.

3.157. It should be mentioned that this second condition for infinite mass of fuel, i.e., when N_m/N_u is zero, has no significance for the present problem because it refers to a system consisting of essentially pure uranium-235 and no moderator. Such a system would not support a thermal-neutron chain reaction, and τ_m no

longer refers to the age of thermal neutrons in the moderator. Actually, as the proportion of uranium-235 is increased, relatively more fissions are caused by neutrons of intermediate and higher energies. The theory of thermal-neutron chain reactions then breaks down and predicts critical masses which are too large.

3.158. Between the two conditions — one corresponding to a very large value of N_m/N_u and the other to a very small value of this ratio — for which the mass of fuel in the critical reactor becomes infinite, there must be a moderator-to-fuel ratio for which the critical mass of fuel is a minimum. The physical basis of this result will be apparent from the following considerations. When N_m/N_u is zero, i.e., z is infinite, the multiplication factor k has its maximum value, which is equal to η, as may be seen from equation (3.145.3). As N_m/N_u is increased, however, k does not change very much, at first. Thus, for a mixture of uranium-235 and graphite, k is 0.99η even when N_m/N_u is as high as 1.4×10^3. This means, then, when N_m/N_u is 1.4×10^3, the critical reactor will have the same nonleakage probability within about 1 per cent, and hence the same size, as when N_m/N_u is almost zero.* The greatly increased moderator-to-fuel ratio in

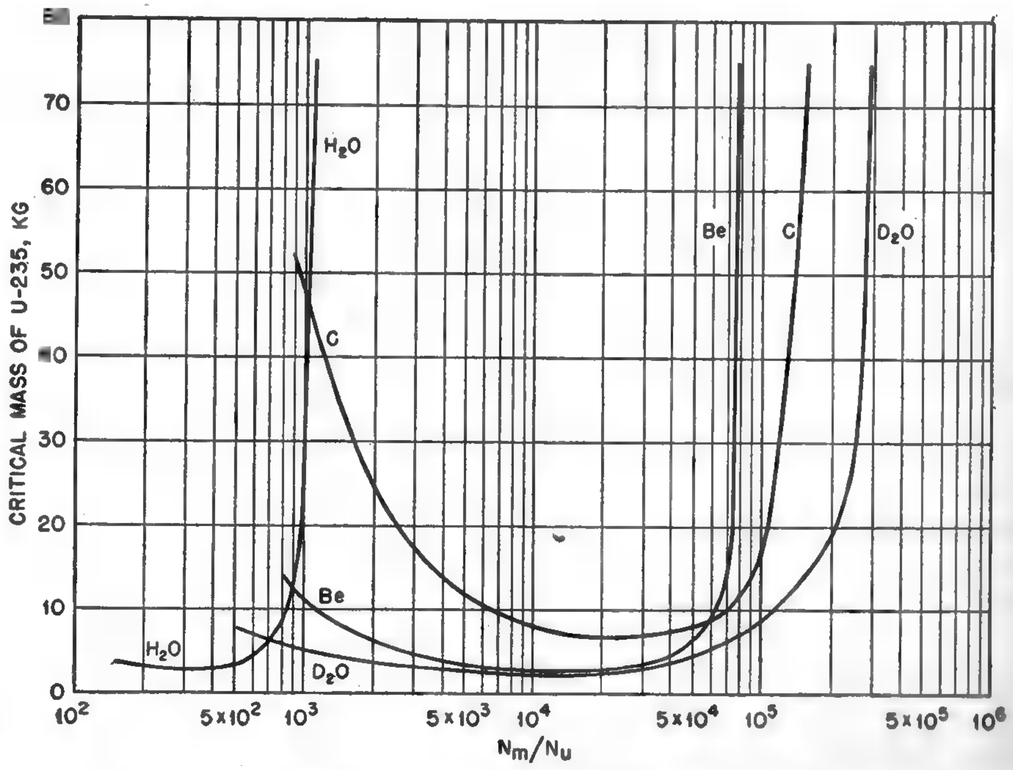


FIG. 3.14. Relationship between critical mass and composition of bare, spherical thermal reactors with pure uranium-235 as fuel

* It is assumed that L^2 and τ are roughly the same in the two cases, although this is not strictly true.

essentially the same total critical volume obviously means a large decrease in the critical mass of fuel as N_m/N_u increases.

3.159. As the ratio N_m/N_u increases still further, k begins to fall off rapidly. Consequently a large increase is necessary in the nonleakage probability, and hence in the reactor size, if the system is to be critical. Beyond a certain point, the critical volume increases more rapidly than N_m/N_u, and then the critical mass of fuel begins to increase with increasing moderator-to-fuel ratio. It is evident, therefore, that there must be certain ratio of the given moderator to fuel for which the critical mass of fuel is a minimum.

3.160. If the reactor composition and geometry, i.e., N_m/N_u and B^2, respectively, are known, it is not a difficult matter to calculate the critical mass of fuel, using the densities of the fuel and moderator. This has been done for systems consisting of uranium-235 as fuel and either heavy water, beryllium, or graphite as moderator, the required data, except for the densities, being obtained from Fig. 3.13. The results plotted in Fig. 3.14* show the dependence of the critical mass of uranium-235 in a bare (spherical) reactor, i.e., one without a reflector, upon the composition, as expressed by the ratio N_m/N_u. The existence of a minimum critical mass is apparent in each case.

3.161. It should be noted that, although some of the arguments presented above have been based on the supposition that pure uranium-235 is the fuel, the general conclusions are applicable to thermal reactors of all types. No matter what the nature of the fuel or the shape of the reactor, there will, in principle, be a particular moderator-to-fuel ratio for which the critical mass of fuel is a minimum.

Water as Moderator

3.162. It was stated in § 3.87 that the continuous slowing down model, which leads to the neutron age concept, cannot be expected to apply when ordinary water is the moderator. This has been confirmed by the experimental fact that the spatial distribution of neutrons about a point source of fast neutrons in water cannot be represented by a simple exponential equation of the form of (3.93.1). The critical equation (3.108.2) does not apply to reactors in which water is moderator, but the expression

$$\frac{k}{(1 + L^2B^2)(1 + L_1^2B^2)(1 + L_2^2B^2)(1 + L_3^2B^2)(1 + L_4^2B^2)} = 1, \quad (3.162.1)$$

which is, in a sense, an extension of equation (3.123.1) may be used. In equation (3.162.1), L is the diffusion length of thermal neutrons in the water-fuel system, and the quantities L_1, L_2, L_3, L_4 are derived from experimental measurements on the spatial distribution of neutrons in water arising from a point source of fast neutrons. The values found are 4.49, 2.45, 2.05, and 1.00 cm, respectively. If the infinite multiplication, k, is known, it is thus possible to calculate

* Based on calculations made by M. C. Edlund.

from equation (3.162.1) the buckling and, hence, the critical size of a reactor in which water is the moderator.

3.163. For a system in which pure uranium-235 is the fuel, the value of k is given by equation (3.145.3); the absorption cross section of the fuel, σ_{au}, is 650 barns and that of the water moderator, σ_{am}, is 0.66 barn. The results of the calculations of B^2 and of the critical mass of fuel, as functions of the moderator-to-fuel ratio, N_m/N_u, have been given in Figs. 3.13 and 3.14, respectively. It should be emphasized that the data refer to a bare reactor, without a reflector. In this case the minimum critical mass of uranium-235 is about 2.7 kg; however, with a good neutron reflector an aqueous solution containing less than 900 grams of uranium-235 can be made critical.

Calculation of Neutron Lifetime

3.164. Since the required information is now available, it is of interest, at this point, to estimate the neutron lifetime in a thermal reactor (§ 3.102). By combining equations (3.102.1) and (3.127.1), the result is

$$l = \frac{1}{v\Sigma_a(1 + L^2B^2)}. \tag{3.164.1}$$

By equation (3.34.1), $\Sigma_a = D/L^2$, where D and L refer to the reactor, i.e., to the fuel-moderator system. As seen in § 3.147, L^2 is equal to $D_m(1 - f)/\Sigma_{am}$, but since the difusion coefficient is actually that in the reactor, D may be substituted for D_m. Upon making these replacements, it is found that Σ_a is equal to $\Sigma_{am}/(1 - f)$, so that equation (3.164.1) becomes

$$l = \frac{1 - f}{v\Sigma_{am}(1 + L^2B^2)}.$$

Further, $1/v\Sigma_{am}$ is the diffusion time (or lifetime) in the moderator alone, as given in Table 3.6; if this is represented by l_m, then

$$l = \frac{l_m(1 - f)}{1 + L^2B^2}.$$

Finally, L^2 in the denominator may be replaced by $L_m^2(1 - f)$, so that

$$l = \frac{l_m(1 - f)}{1 + L_m^2B^2(1 - f)},$$

from which the lifetime of thermal neutrons in the reactor can be readily estimated.

Example Calculate the lifetime of thermal neutrons in the reactor considered in the example following § 3.150.

Since the moderator is beryllium, l_m is 4.3×10^{-3} sec (Table 3.6) and L_m is 23.6 cm. By equation (3.145.2), the value of $1 - f$ is given by $1/(z + 1)$, which is 1/8.2, i.e., 0.12, for the reactor under consideration. For this reactor, too, B^2 was found to be 3.8×10^{-3} cm^{-2}. Consequently,

$$l = \frac{(4.3 \times 10^{-3})(0.12)}{1 + [(23.6)^2(3.8 \times 10^{-3})(0.12)]} = 4.2 \times 10^{-4} \text{ sec.}$$

The required thermal-neutron lifetime is thus 4.2×10^{-4} sec. (The main reason why this is so much smaller than in the moderator alone is because of the greater probability of neutron absorption in the fuel-moderator mixture.)

HETEROGENEOUS NATURAL URANIUM REACTORS*

Effective Resonance Integral

3.165. A reactor with natural uranium as fuel can be made critical if, instead of using a uniform mixture of fuel and moderator, the system is constructed as a heterogeneous lattice consisting of lumps (or rods) of natural uranium arranged in a matrix of graphite. Although such an arrangement results in a decrease in the thermal utilization, this is more than offset by the increase in the resonance escape probability. With the proper choice of dimensions and spacing of the fuel rods the infinite multiplication factor can be made to exceed unity, provided that neutron-absorbing impurities are kept to a minimum.

3.166. The increase in the resonance escape probability resulting from lumping of the fuel is due mainly to the decrease in the effective resonance integral, as compared with that for a uniform system of the same composition. The physical interpretation of this fact is that, because of the high resonance peaks of uranium-238, neutrons which are slowed down to resonance energies in the moderator are largely absorbed in the outer layers of the uranium lump. The resonance flux within the lump is, consequently, sharply depressed, and as a result the resonance absorption is decreased compared with the thermal absorption. By postulating that the effective absorption cross section for a lumped system can be separated into two parts, namely, volume absorption and surface absorption, it has been found that the experimental values of the effective resonance integral (per atom) for natural uranium at ordinary temperatures can be expressed approximately by

$$\int (\sigma_{au})_{\text{eff}} \frac{dE}{E} = 9.25 + 24.7 \frac{S}{M} \text{ barns,} \tag{3.166.1}$$

where S is the surface area of the uranium lumps in cm^2 and M is the mass in grams.

3.167. It is evident from equation (3.166.1) that the effective resonance integral will increase as the lumps are made smaller, i.e., as S/M is increased. For a given fuel-to-moderator ratio, the resonance escape probability will thus decrease as the system approaches uniformity. For lumps consisting of long

* For a general discussion of the theory of heterogeneous reactors, see S. Glasstone and M. C. Edlund, *op. cit.*, Chapter IX. Applications of the theory to criticality calculations of natural uranium-graphite systems are given by E. A. Guggenheim and M. H. L. Pryce, *Nucleonics*, **11**, No. 2, 50 (1953); see also, R. O. Fornaguera, *et al.*, *Chem. Eng. Prog. Symposium Series*, No. 12, **50**, 88 (1954).

rods or spheres of uranium metal of 1 cm radius, the value of S/M is roughly 0.1, and so the effective resonance integral is approximately 12. This may be compared with an experimental value of 100 for the uniform mixture containing 0.0025 part of uranium metal to one of graphite which gives the maximum value of the product pf in Table 3.8. The much smaller resonance integral for the heterogeneous arrangement means that the resonance escape probability can be much closer to unity, for the same over-all composition.

3.168. It should be mentioned that the decrease in the effective resonance integral due to lumping of the fuel is to be attributed, ultimately, to the fact that the uranium-238 resonances are high and narrow (see Fig. 2.18). There is thus a considerable probability that neutrons having energies at, or very close to, the resonances are absorbed in the outer layers of the lump. All others with energies above thermal will have a high probability of passing through the lump and re-entering the moderator, where they are slowed down. If the uranium resonances had been broad and low, the absorption in the outer layers would be less, and uranium nuclei in the interior of the lump would be exposed to a relatively higher resonance flux. Further, because of the wider range of neutrons absorbed, the proportion returning to the moderator would be decreased. Both these effects would result in a decrease in the resonance escape probability.

3.169. Another, although less important, increase in the resonance escape probability, amounting to a few per cent only, in a lumped system arises from the fact that some of the neutrons will be slowed down through the resonance region to thermal energies in the moderator without coming into contact with fuel nuclei. Such a situation would not be possible in a uniform mixture of fuel and moderator.

Resonance Escape Probability

3.170. The expression for the resonance escape probability given earlier, i.e., equation (3.135.3), is applicable to a uniform mixture where both fuel and moderator are exposed to the same neutron flux (see § 3.78). For a heterogeneous system, the equation must be modified and now takes the approximate form

$$p(E) \approx \exp\left[-\frac{N_u V_u \phi_u}{\xi \Sigma_{sm} V_m \phi_m} \int (\sigma_{au})_{\text{eff}} \frac{dE}{E}\right], \tag{3.170.1}$$

where V_m and V_u are the volumes of moderator and fuel in the lattice, and ϕ_m and ϕ_u are the *average* values of the *resonance* flux in the moderator and in the interior of the fuel lump, respectively. The ratio ϕ_m/ϕ_u is called the *disadvantage factor* for resonance neutrons.

3.171. For a given ratio of moderator to fuel, i.e., V_m/V_u constant, the depletion of the resonance flux, and ϕ_m/ϕ_u, will be greater, the larger the radius of the fuel lumps. Hence for a specified composition, the value of $p(E)$ will increase with increasing size of the lumps because of the gain in the disadvantage

factor ϕ_m/ϕ_u. This will be in addition to the increase due to the change in the effective resonance integral. On the other hand, for lumps of specified size, so that ϕ_m/ϕ_u is approximately constant, the resonance escape probability will increase as V_m/V_u increases. The general conclusions are therefore that the resonance escape probability will increase (1) with increasing radius of the fuel lump (or rod) at constant moderator-fuel ratio, and (2) with increasing value of the moderator-fuel ratio, V_m/V_u, at constant radius of the fuel lump (or rod).

3.172. The calculation of the resonance escape probability in any particular case requires a knowledge of the resonance disadvantage factor ϕ_m/ϕ_u, and this is determined from the physical properties and geometry of the heterogeneous lattice. For this purpose a quantity f_r, called the *resonance utilization*, because of its analogy to the familiar thermal utilization (§ 3.133), is introduced. It is defined as the ratio of the neutrons absorbed by the fuel in the resonance region to the total number of resonance neutrons produced. As a result, equation (3.170.1) can be transformed into

$$p(E) \approx \exp\left[-\frac{N_u \Sigma_{am}}{\xi \Sigma_{sm} \Sigma_{au}} \cdot \frac{f_r}{1 - f_r} \int (\sigma_{au})_{\text{eff}} \frac{dE}{E}\right].$$

The evaluation of f_r is based on the relationship

$$\frac{1}{f_r} = \frac{V_m \Sigma_{am}}{V_u \Sigma_{au}} F + E, \tag{3.172.1}$$

where F and E are known functions of the diffusion length of resonance neutrons in fuel and moderator, and of the reactor geometry, i.e., the over-all shape of

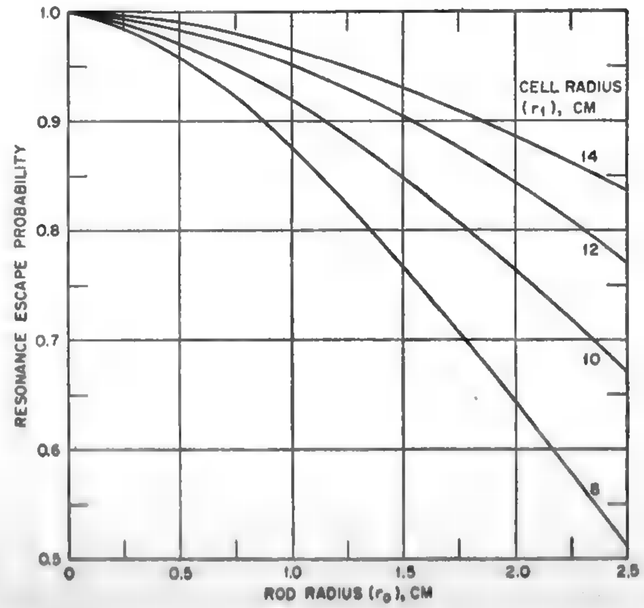


Fig. 3.15. Resonance escape probability in heterogeneous natural uranium-graphite systems

the system and the size of the fuel rods (or lumps) and their spacing in the lattice. It should be noted that the cross sections refer to resonance neutrons, and the value for the fuel (natural uranium) is obtained from the effective resonance integral given earlier.

3.173. The results of some calculations of the resonance escape probability in various lattices consisting of a square array of rods of natural uranium in a matrix of graphite are shown in Fig. 3.15. The radius of the rod is represented by r_0 and the symbol r_1 indicates what is called the *cell radius;* the latter is the radius of a circle having the same area as the unit square upon which the lattice is based. The value of V_m/V_u for the array with cylindrical fuel rods is equal to $(r_1^2 - r_0^2)/r_0^2$, since this is the ratio of the geometrical cross sections (and volumes) of the graphite and uranium cylinders in a unit cell.

Thermal Utilization

3.174. As stated earlier, lumping of the fuel results in a decrease in the thermal utilization. Disregarding absorption of neutrons by impurities, equation (3.133.1), for a heterogeneous system, may be written as

$$f = \frac{\Sigma_{au} V_u \phi_u}{\Sigma_{au} V_u \phi_u + \Sigma_{am} V_m \phi_m} = \frac{1}{1 + \dfrac{\Sigma_{am}}{\Sigma_{au}} \cdot \dfrac{V_m \phi_m}{V_u \phi_u}}, \tag{3.174.1}$$

where ϕ_m and ϕ_u are now the *average* values of the *thermal*-neutron flux in moderator and fuel, respectively; the macroscopic absorption cross sections also, of course, refer to thermal neutrons. For a given volume ratio of moderator to

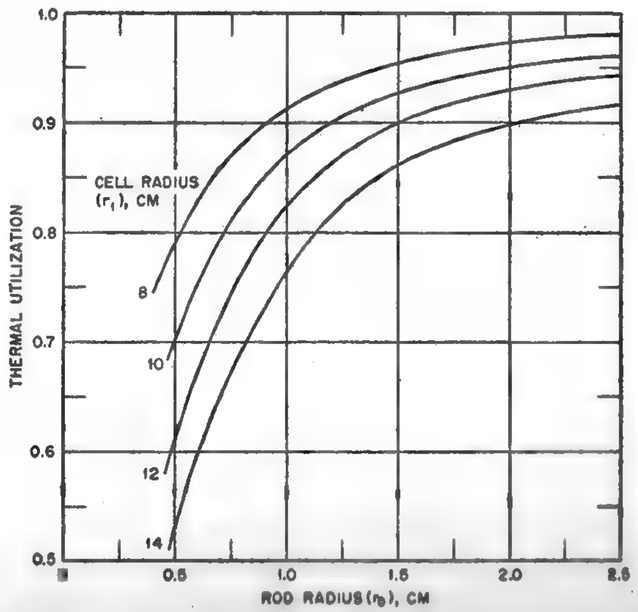


Fig. 3.16. Thermal utilization in heterogeneous natural uranium-graphite systems

fuel, the thermal utilization will decrease as the thermal disadvantage factor ϕ_m/ϕ_u increases. The effect of changing the radius of the fuel lump is thus the reverse of that on the resonance escape probability. Similarly, for a lump of given radius, an increase in V_m/V_u would cause a decrease in the thermal utilization, whereas the resonance escape probability would be increased. It is seen, therefore, that the thermal utilization should increase (1) with decreasing radius of lump (or rod) at constant composition, and (2) with decreasing V_m/V_u at constant radius of fuel lump (or rod).

3.175. In order to evaluate the thermal utilization for any particular heterogeneous fuel-moderator lattice, its reciprocal is written in a form exactly equivalent to equation (3.172.1), except that Σ_{am} and Σ_{au} are now thermal-neutron absorption cross sections. The functions F and E are the same as before, with the diffusion lengths also those for thermal neutrons. The results of some calculations for a square lattice of uranium rods in graphite are given in Fig. 3.16.

Fast-fission Factor

3.176. Since η may be regarded as known for natural uranium (§ 3.130), there remains only the fast-fission factor ϵ to be determined in order that the infinite multiplication factor may be evaluated for a given lattice. The method of cal-

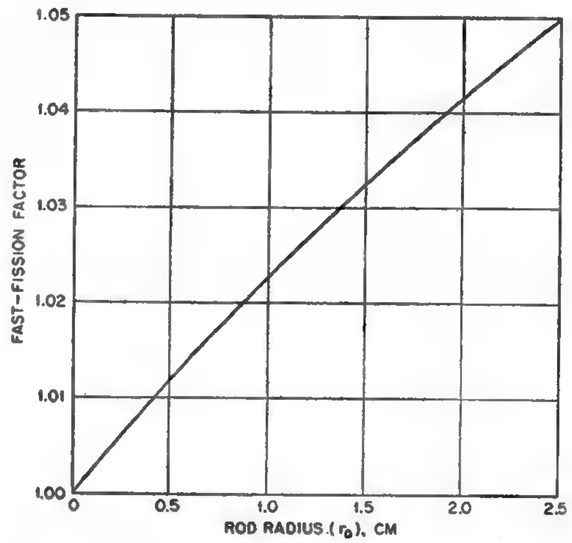


Fig. 3.17. Fast-fission factor in natural uranium fuel rods

culation is based on probability theory and requires a knowledge of fission, nonfission capture, and inelastic and elastic scattering cross sections for fast neutrons in the fuel. The values of ϵ increases with the probability that a primary fission neutron will make a collision of some kind inside the fuel rod (or lump) in which it was created. As may be expected, this probability increases with the total neutron cross section of the fuel material and with the radius of

the rod, and consequently so also does the fast-fission factor. Lumping of the fuel thus provides some advantage, although a relatively small one, due to the increase in the fast-fission factor. The dependence of ϵ upon the radius of a cylindrical fuel rod of natural uranium is indicated in Fig. 3.17. In the Oak Ridge (X-10) graphite-moderated reactor and in the heavy-water moderated CP-3, the rod radius is about 1.4 cm, so that ϵ is 1.030.

Determination of Optimum Lattice

3.177. An examination of Figs. 3.15 and 3.16 shows that the conditions which favor an increase in the resonance escape probability are just those which cause the thermal utilization to decrease, and vice versa. The problem in the construction of a heterogeneous lattice is to determine the particular arrangement of fuel and moderator that gives a maximum value of the product pf. The procedure used is to consider a number of fuel-moderator lattices, with various rod radii and lattice spacings (cell radii), and to calculate the values of p, f, and ϵ for each lattice. Then, using the known value of η, the corresponding infinite multiplication factors, k, can be determined.

Example In the Oak Ridge natural uranium-graphite reactor the cylindrical fuel rods of 1.1 in. diameter are arranged in a square lattice with a pitch of 8 in. Neglecting the effect of the aluminum cladding of the rods and the air space around them, evaluate the infinite multiplication factor of the system.

Since η for natural uranium is known to be 1.3_2 the problem involves the calculation of p, f, and ϵ; these will be considered in turn. To calculate p, equation (3.170.1) will be used and the values of the various component factors will first be determined. The calculation of the resonance disadvantage factor ϕ_m/ϕ_u is lengthy and it will be assumed to be unity, since experience has shown that this is a fair approximation for the given lattice. The value of V_m/V_u, as seen in § 3.173, is given by $(r_1^2 - r_0^2)/r_0^2$, where πr_1^2 is equal to the area of a unit square, i.e., 64 in.^{2}, so that $r_1^2 = 64/\pi$ in.^{2}; r_0, the rod radius, is 0.55 in. Hence,

$$\frac{V_m}{V_u} = \frac{r_1^2 - r_0^2}{r_0^2} = \frac{(64/\pi) - (0.55)^2}{(0.55)^2}$$
$$= 66.5.$$

The quantity N_u/Σ_{sm} may be written as $N_u/N_m\sigma_{sm}$, where σ_{sm} for graphite is 4.8 barns. In order to determine N_u/N_m it is found from equation (2.113.1) that

$$\frac{N_u}{N_m} = \frac{\rho_u A_m}{\rho_m A_u},$$

where the ρ's and A's are the densities and atomic weights, respectively, of the fuel and moderator. For uranium ρ_u is 19.0 g/cm^3 and A_u is 238, whereas for graphite ρ_m is 1.62 g/cm^3 and A_m is 12.0, so that

$$\frac{N_u}{N_m} = \frac{(19.0)(12.0)}{(1.62)(238)} = 0.592.$$

Since ξ for graphite is known to be 0.158, there remains only the effective resonance integral to be evaluated from equation (3.166.1). Neglecting the surface area of the

ends of the rods, since this is a small fraction of the whole in a long cylindrical arrangement, the ratio of surface to mass, i.e., S/M, for a cylindrical fuel rod of radius r_0 is given by

$$\frac{S}{M} = \frac{2\pi r_0}{(\pi r_0^2)\rho_u} = \frac{2}{r_0\rho_u},$$

with r_0 expressed in cm and ρ_u in g/cm^3. The rod diameter is 1.1 in. and so the radius r_0 is 1.40 cm; consequently

$$\frac{S}{M} = \frac{2}{(1.40)(19.0)} = 0.0752$$

and, by equation (3.166.1),

$$\int (\sigma_{au})_{\text{eff}} \frac{dE}{E} = 9.25 + (24.7)(0.0752)$$
$$= 11.1 \text{ barns.}$$

Upon substituting the foregoing results into equation (3.170.1) it is found that

$$p = \exp\left[-\frac{(0.592)(11.1)}{(0.158)(4.8)(66.5)(1.0)}\right]$$
$$= e^{-0.139} = 0.87_8.$$

The calculation of f is made by means of equation (3.174.1) and here

$$\frac{\Sigma_{am}}{\Sigma_{au}} = \frac{N_m\sigma_{am}}{N_u\sigma_{au}},$$

where the σ's are the thermal absorption cross sections, i.e., 0.0045 barn for graphite and 7.42 barns for natural uranium. Since N_u/N_m has already been found to be 0.592, it is seen that

$$\frac{\Sigma_{am}}{\Sigma_{au}} = \frac{N_m\sigma_{am}}{N_u\sigma_{au}} = \frac{0.0045}{(0.592)(7.42)}$$
$$= 1.02 \times 10^{-3}.$$

The value of V_m/V_u is 66.5, as seen earlier, and so there remains only the thermal disadvantage factor, ϕ_m/ϕ_u, to be determined. For fuel-moderator arrangements of practical interest this may be taken as approximately 1.5, which means that the average thermal-neutron flux in the fuel rod is about two thirds of the average value in the moderator. It is then found from equation (3.174.1) that

$$f = \frac{1}{1 + (1.02 \times 10^{-3})(66.5)(1.5)} = \frac{1}{1.102}$$
$$= 0.90_8.$$

From Fig. 3.17, the value of ϵ for a rod 1.40 cm radius is 1.03_0, so that the infinite multiplication factor is given by

$$k = \eta\epsilon pf = (1.3_2)(1.03_0)(0.87_8)(0.90_8) = 1.08_4.$$

(The actual value of k for the system under consideration is about 1.065, the difference being partly due to the neglect of neutron absorption in the aluminum cladding of the

fuel elements and in the cooling air, and also to some extent to the approximate nature of the calculation given above.)

3.178. Some indication of the results obtained from a series of calculations similar to that just described may be obtained from the data in Table 3.9.* These refer to a natural uranium-graphite lattice, with cylindrical fuel rods of 1.25 cm radius located in cells of various radii. Allowance is made in each case for an aluminum cladding 0.115 cm thick and a 2-cm air space surrounding each fuel rod. The value of η used in the calculations is 1.308 and that of ϵ for the given rods was taken to be 1.028.

TABLE 3.9. PROPERTIES OF HETEROGENEOUS NATURAL URANIUM-GRAPHITE LATTICES

Cell Radius (cm)	f	p	k
10	0.907	0.866	1.055
11	0.888	0.905	1.0625
12	0.8765	0.9085	1.060
13	0.846	0.923	1.0495

3.179. It is apparent that, for a specified rod radius, the value of k passes through a maximum as the cell radius, i.e., the pitch of a square lattice, is increased. This maximum generally occurs when the values of p and f, one of which decreases regularly while the other increases, are almost equal.

3.180. If a particular cell radius is chosen and the calculations performed for different radii of the fuel rods, somewhat similar results are obtained. With increasing rod radius the values of f increase, whereas those of p decrease, so that k has a maximum for a particular radius. By combining the various data it is thus possible to choose the best dimensions for the reactor. It was on the basis of such calculations that the lattices of the natural uranium-graphite reactors were designed, e.g., rod radius 1.4 cm (about 1.1 in. diameter) and 11.5 cm cell radius (about 8 in. pitch in square lattice).†

Heavy-Water-Moderated Heterogeneous Reactors

3.181. If heavy water is used instead of graphite as moderator in a heterogeneous reactor with natural uranium as fuel, appreciably higher values of the infinite multiplication factor are possible. Although Σ_{sm} for heavy water is not greatly different from that of graphite (see Table 8.9), the fact that ξ for the former is 0.51, compared with 0.158 for the latter, means that, according to equation (3.170.1), the resonance escape probability, under equivalent conditions, will be much closer to unity when heavy water is the moderator. For

* E. A. Guggenheim and M. H. L. Pryce, *loc. cit.*

† For an analytical procedure, see I. C. Atkinson and R. L. Murray, *Nucleonics*, **12**, No. 4, 50 (1954).

instance, in the system described in the example following § 3.177, the value of p would be increased to about 0.96 if the graphite were replaced by heavy water. The reason for the increase in the resonance escape probability is that the average energy change per collision is greater than in graphite, so that the neutrons are more likely to be slowed down past the resonance region in a collision.

3.182. In addition to the increase in the resonance escape probability in heavy water, there is also an increase in the thermal utilization. This is due to the smaller macroscopic absorption cross section than for graphite. It will be seen from equation (3.174.1) that a decrease in Σ_{am} will result in an increase in f, which may well be as high as 0.95 if the other terms remain essentially unchanged. In other words, by the use of heavy water a smaller proportion of the neutrons are lost by capture in the moderator. Taking η as 1.32 and ϵ as 1.03, it is possible for k to be about 1.25 or 1.20, at least, if allowance is made for extraneous neutron capture in the fuel-element cladding, etc. As a result of the significant increase in the infinite multiplication factor, a heavy-water moderated reactor can have a smaller critical size and require less fuel than one moderated by graphite, in spite of the smaller diffusion length in the latter.

3.183. Because heavy water is a better moderator than graphite, a smaller ratio of moderator to fuel is necessary. Consequently the fuel rods can be more closely spaced when heavy water is the moderator. This was the case in the CP-3 reactor, for example, where the 1.1-in. diameter rods of natural uranium were arranged $5\frac{3}{8}$ in. apart, as compared with 8 in. in graphite. The decrease in the value of V_m/V_u causes a small decrease in the resonance escape probability, but there is some compensating increase in the thermal utilization, so that the over-all effect on k is small. The reduction in the quantity of the expensive moderator (and fuel) is, however, important. Thus a bare reactor moderated by heavy water and having natural uranium for the fuel requires about 3 to 4 tons of uranium and 7 tons of moderator for criticality, as compared with something like 50 tons of uranium and 280 tons of moderator when graphite is used.

Ordinary Water as Moderator: Slightly Enriched Fuel

3.184. Ordinary water is a better moderator than heavy water in the respect that both ξ and Σ_{sm} are larger. As a result it is to be expected that a higher resonance escape probability can be achieved in ordinary water, although a value as high as 0.96 is possible in heavy water, so that the advantage of ordinary water in this connection is not significant. However, because of the much larger macroscopic absorption cross section for thermal neutrons in ordinary water (0.022 cm^{-1}, compared to 8.5×10^{-5} in heavy water and 3.7×10^{-4} cm^{-1} in graphite) there is an inevitable large decrease in the thermal utilization. In fact, it is doubtful whether f can exceed 0.75 when p is up to 0.96. The maximum value of the multiplication factor in any arrangement of natural uranium

in ordinary water is probably less than unity, so that it appears to be impossible to obtain a critical system.*

3.185. The problem of attaining criticality with water as moderator can be solved by the use of enriched fuel. The treatment of mixtures containing highly enriched fuel material, e.g., almost pure uranium-235, was given earlier, and a brief discussion will be presented here of slightly enriched fuels. Such fuel materials used in conjunction with ordinary water as moderator are likely to be of interest for power production, as will be seen later. The most important effect of enrichment in uranium-235 is the increase in η, as is apparent from the results in Table 3.10. It will be noted that even a small degree of enrichment can produce a marked increase in η; thus the change from 0.71 per cent of uranium-235 (in natural uranium) to 2.0 per cent already increases η roughly halfway toward the value for pure uranium-235.

TABLE 3.10. MULTIPLYING PROPERTIES OF ENRICHED FUELS

Uranium-235 (*per cent*)	η	k (*approx.*)
0.71	1.3_2	0.97
1.00	1.4_8	1.1_1
1.20	1.5_6	1.1_7
1.50	1.6_5	1.2_4
2.00	1.7_3	1.3_1
100	2.1	1.6

3.186. Besides causing an appreciable increase in η, enrichment of the fuel could decrease the effective resonance integral, and so increase the resonance escape probability, due to the smaller proportion of uranium-238 present. However, this will probably be more than offset by the effect of the smaller spacing between the fuel elements in ordinary water, as compared with other moderators, and the resulting decrease in V_m/V_u. As seen from equation (3.170.1), such a change will cause p to decrease. On the other hand, equation (3.174.1) shows that a decrease in V_m/V_u, i.e., a decrease in the proportion of the neutron-absorbing moderator, will be accompanied by an increase in the thermal utilization. Since p and f change in opposite directions, the product will be very roughly constant, and on this basis the approximate maximum multiplication factors recorded in Table 3.10 have been estimated. The actual values will vary with the size of the fuel rods (or plates) and their spacing, and they will be decreased by parasitic neutron capture in cladding, etc. Nevertheless, the data indicate that a critical system with ordinary water as moderator can be obtained with a fuel containing 1 per cent of uranium-235. A further increase in the enrichment results in an increase in k, so that reactors of smaller size are possible.

* Cf. R. Persson, *Nucleonics*, **12**, No. 10, 26 (1954).

3.187. It may be remarked that the main purpose of fuel lumping, which is to increase the resonance escape probability, is less important for water than for other moderators, especially when the spacing between the fuel elements is relatively small. In these circumstances N_m/N_u is small and so also is Σ_{sm}/N_u; the effective resonance integral for the homogeneous system, as expressed by equation (3.136.1), is then relatively small and there is not a great deal to be gained by making the system heterogeneous. Because of this circumstance an arrangement of closely spaced fuel elements of enriched uranium in water as moderator can be treated, for purposes of calculation, as being homogeneous, although it is actually heterogeneous. It should be noted, however, that whereas the fast-fission factor in a homogeneous system is close to unity, it can be appreciably larger in a uranium-water lattice.

Calculation of Critical Size

3.188. The critical equation (3.108.2) is strictly applicable to homogeneous systems. However, if the fuel rods (or plates) are not very close together, the over-all neutron flux and slowing down density distributions in a heterogeneous system, apart from variations within the fuel itself, will be similar to that in a homogeneous reactor. The critical equation for the heterogeneous arrangement will then have the same form as equation (3.108.2). Natural-uranium reactors are inevitably large, as k is not much greater than unity, even when heavy water is the moderator, and so equation (3.123.3) may be used. The buckling of the system is then given by

$$B^2 = \frac{k-1}{M^2},$$

where $M^2 = L^2 + \tau$.

3.189. Since k may be regarded as known, the calculation of B^2, and thus of the critical size, involves the determination of the appropriate values of L^2 and τ. It can be shown that L^2 is equal to $L_m^2(1-f)$, which is of the same form as for a homogeneous system (§ 3.147), with f having the value appropriate to the chosen lattice. The thermal-neutron age, τ, may be expected to be somewhat larger than for the moderator alone, because uranium contributes little to the slowing down of neutrons by elastic collisions. In this respect, the fuel lumps consequently behave as voids. However, this tendency to increase τ is opposed by the inelastic scattering of high-energy neutrons by uranium nuclei. The value of τ for the given lattice can be calculated, but as a rough approximation the age of thermal neutrons in a heterogeneous lattice may be taken to be about 10 per cent larger than for the pure moderator.

3.190. With k and M^2 available, the magnitude of B^2 in the bare critical reactor can be evaluated. As before, this is set equal to the appropriate expression for the buckling (Table 3.7), and the dimensions of the critical reactor can then be calculated.

Example From the data already given, estimate the mass of natural uranium required to make a bare cubical reactor, with graphite as moderator, just critical.

The best value for k is probably 1.065, and then f will be 0.889 (Table 3.9). For graphite, L_m^2 is 2500 cm^2 (Table 3.1), and so L^2 for the reactor, which is equal to $L_m^2(1-f)$, as stated above, is 280 cm^2. The thermal age, τ, in graphite is 350 cm^2 (Table 3.4), but in the reactor it is somewhat larger, so that 400 cm^2 is a good estimate. Consequently, $M^2 = L^2 + \tau = 280 + 400 = 680$ cm^2, and

$$B^2 = \frac{k-1}{M^2} = \frac{(1.065-1)}{680} = 9.6 \times 10^{-5} \text{ cm}^{-2}.$$

For a cubical reactor, $B^2 = 3(\pi/a)^2$, where a is the edge length: hence,

$$a^2 = 3\left(\frac{\pi}{B}\right)^2 = \frac{(3)(3.14)^2}{9.6 \times 10^{-5}} = 31 \times 10^4 \text{ cm}^2.$$

The edge length, a, is thus 560 cm, or nearly 19 ft. This includes the extrapolation distance, but as it is only about 2 cm it may be ignored.

Assuming the lattice to consist of a square array with a pitch of 8 in., the cross section of a unit cell is 64 in.^{2}, or 410 cm^2. Since the area of a face of the cube, which is equal to a^2, is 31×10^4 cm^2, it is seen that there must be $31 \times 10^4/410 = 750$ unit cells. Each cell contains the equivalent of a single rod of fuel, and so there are 750 fuel rods.

The radius of a fuel rod may be taken to be 1.4 cm and its length is equal to that of the cube edge, i.e., 560 cm. The total volume of the 750 rods is

$$\text{Volume of fuel} = (\pi)(1.4)^2(560)(750) = 26 \times 10^5 \text{ cm}^3.$$

The density of uranium metal is 19 g/cm^3, and so the total weight of uranium in the bare critical reactor is

$$\text{Weight of fuel} = (26 \times 10^5)(19) = 4.9 \times 10^7 \text{ grams} \approx 54 \text{ tons}.$$

(It will be seen later that, by using a reflector, the critical mass of fuel can be reduced to little more than half this amount.)

EXPERIMENTAL DETERMINATION OF CRITICAL SIZE

The Critical Assembly

3.191. Confirmation of the calculation of the critical size of a reactor, either homogeneous or heterogeneous, is obtained by experiment. The method of the critical assembly can be used for reactors of all sizes, but it is more suited to small (enriched) reactors. A system consisting of fuel and moderator, in the desired proportions and shape, is gradually built up until it approaches the critical dimensions. At the center of the assembly is placed a source, emitting about 10^7 neutrons/sec; this is called the primary source. As a result of the fissions induced in the fuel material, there is a multiplication of the primary source neutrons. Even though the reactor is less than the critical size, a steady state of the neutron flux will be attained, provided the source is present. The (subcritical) multiplication is then defined as the ratio of the total thermal-neutron flux, due to both the primary source and the fissions, to the flux due to the source only, i.e., in the same medium in the absence of fissionable material.

3.192. If Q thermal neutrons are obtained from the source, these will produce Qk_{eff} thermal neutrons at the end of one generation, Qk^2_{eff} at the end of two generations, and so on. The effective multiplication factor, k_{eff}, for the particular assembly under consideration depends on its size as well as its composition and arrangement. The steady-state multiplication of the neutrons in the system can then be represented by

$$\frac{Q + Qk_{\text{eff}} + Qk_{\text{eff}}^2 + \cdots}{Q} = \frac{1}{1-k_{\text{eff}}}, \qquad (3.192.1)$$

provided $k_{\text{eff}} < 1$, i.e., the assembly is subcritical. As the size of the assembly is increased and approaches critical, k_{eff} tends to unity, and the multiplication becomes infinite.

3.193. The multiplication can be determined experimentally by measuring the thermal-neutron flux within the assembly at a certain distance from the source. The measurement is first made in the absence of the fuel, i.e., with source and moderator only, and then repeated at the same position with fuel present. The ratio of the two values gives the required multiplication at the given point for a particular size of the assembly. The observations are repeated for assemblies of various sizes approaching the critical.

3.194. In order to determine the actual critical size the reciprocal of the multiplication is plotted against the size of the assembly, as in Fig. 3.18. The plot is then extrapolated to zero value of the reciprocal multiplication, i.e., infinite multiplication, to give the critical size of the system. By making observations at two (or more) different positions, two (or more) sets of data are obtained, and the curves should extrapolate to the same point, as shown in the figure. The size of the critical reactor can thus be determined from measurements with subcritical assemblies. A final check can then be made by constructing the critical reactor and showing that a chain reaction can be maintained in the absence of an extraneous source.

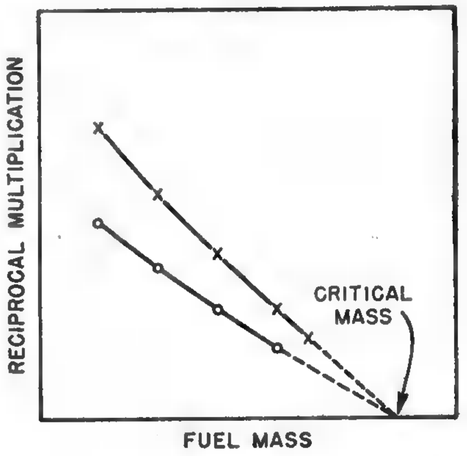


Fig. 3.18. Determination of critical mass

3.195. In view of the statement based on equation (3.192.1) that the multiplication is infinite when k_{eff} is unity, it might appear that an exactly critical reactor, i.e., one for which $k_{\text{eff}} = 1$, would be unstable. In a sense, this is true if a primary source is present. For stability, k_{eff} must be less than unity, but the difference from unity may be only infinitesimal, as will be seen from the following calculations. A good primary source might be one emitting 10^7

neutrons/sec; in a reactor of volume 10^6 cm^3, this would represent 10 neutrons/(cm^3)(sec).

3.196. The thermal-neutron flux in a critical reactor might be 2×10^{13} neutrons/(cm^2)(sec), and taking the mean thermal velocity as 2×10^5 cm/sec (§ 1.54), the neutron density would be 10^8 neutrons/cm^3. The average lifetime of a thermal neutron, e.g., in a natural uranium-graphite system, is about 10^{-3} sec. Hence neutrons are produced in the reactor at the rate of 10^{11} neutrons/(cm^3)(sec). In other words, the 10 primary source neutrons are multiplied up to 10^{11} as a result of the fission chain in the fuel. The neutron multiplication, in this case, is consequently 10^{10}.

3.197. If this result is inserted into equation (3.192.1), it is seen that k_{eff} is $1 - 10^{-10}$, which is indistinguishable from unity for all practical purposes. Therefore, while it is true that if a primary neutron source is present,* as it frequently is, k_{eff} cannot actually be unity, if the reactor is to be stable, the departure from unity is so small as to be negligible if the reactor is operating at an appreciable power level. In the complete absence of any primary source, which is an essentially impossible situation, a steady state could be maintained only if k_{eff} were precisely unity.

The Exponential Experiment†

3.198. In an exponential (pile) experiment, a subcritical assembly is constructed having exactly the same composition and structure as the proposed (large) reactor, but with linear dimensions about one third or one fourth of those expected for the critical system. If a primary source is present, a steady state can be attained in the subcritical assembly, as described above. The flux distribution in such an assembly is not the same as that in a critical reactor. Nevertheless, it can be shown that if the subcritical system is relatively large, as would be the case for a natural-uranium assembly, the over-all (or large-scale) thermal-neutron flux distribution, at a distance from boundaries and from the source, can be represented fairly well by

$$\nabla^2\phi + B^2\phi = 0, \tag{3.198.1}$$

where B^2 is the buckling of the given fuel-moderator system when critical. The value of B^2 is determined from measurements of the thermal flux distribution in the regions of the large subcritical assembly where equation (3.198.1) may be expected to apply.

3.199. The subcritical assembly, e.g., in the form of a rectangular parallelepiped or a cylinder, is constructed upon a base from which emerges a constant supply of thermal neutrons. The thermal-neutron source may consist of a

* Such a source is used in some reactors to simplify start-up (§ 6.161), but, in any case, spontaneous fission and cosmic rays generally provide a primary source of low intensity.

† For theory and experimental details, see S. Glasstone and M. C. Edlund, *op. cit.*, p. 281, *et seq.*, E. C. Campbell, *et al.*, Report AECD-3169; E. R. Cohen, *Chem. Eng. Prog. Symposium Series*, No. 12, 50, 72 (1954).

structure of graphite and uranium within which is placed an ordinary neutron source, or it may be part of an existing thermal reactor. If z is the vertical distance of a point from the bottom of the assembly, then theoretical considerations show that the thermal-neutron flux in the z-direction is given by

$$\phi(z) = Ce^{-\gamma z}, \tag{3.199.1}$$

where C is a constant. This relationship, which holds at appreciable distances from the source and boundaries, means that, in the experimental arrangement under consideration, the neutron flux along any line parallel to the (vertical) z-axis falls off in an exponential manner. It is for this reason that the system has been called an "exponential pile."

3.200. According to equation (3.199.1) the plot of ln ϕ against z should be a straight line of slope $-\gamma$, so that the latter can be determined from experimental measurements of the thermal flux at various points along a z-axis, not too near the boundaries of the assembly. The importance of this result lies in the fact that $-\gamma^2$ constitutes one of the terms in the expression for the buckling (Table 3.7). Thus, for an exponential assembly having the form of a rectangular parallelepiped, with dimensions a and b in the x- and y-directions,

$$B^2 = \left(\frac{\pi}{a}\right)^2 + \left(\frac{\pi}{b}\right)^2 - \gamma^2.$$

For a cylinder of radius R,

$$B^2 = \left(\frac{2.405}{R}\right)^2 - \gamma^2,$$

the value of γ being obtained from the flux variation in the vertical direction. Hence, if γ is determined, as just described, the value of B^2 can be obtained. The dimensions of the critical reactor, having the same structure and composition as the experimental assembly, can then be calculated.

Example Estimate the minimum critical mass of fuel required for a bare cylindrical reactor with heavy water as moderator in which natural uranium fuel rods of 1.4-cm radius are arranged vertically in a square array of $5\frac{1}{2}$-in. pitch, from the fact that in an exponential experiment with a cylinder of 56-cm radius the value of γ, derived from the vertical variation of the thermal-neutron flux, was found to be 28.8×10^{-3} cm^{-1}.

The value of γ^2 is 825×10^{-6} cm^{-2} and so, for a cylinder,

$$B^2 = \left(\frac{2.405}{R}\right)^2 - \gamma^2 = \left(\frac{2.405}{56}\right)^2 - 825 \times 10^{-6}$$
$$= 1.0 \times 10^{-3} \text{ cm}^{-2},$$

and hence B is 3.2×10^{-2} cm^{-1}. (The extrapolation length should be added to the radius of the cylinder to obtain R, but since the reactor is fairly large, its omission will not greatly affect the results which are, in any event, approximate.)

The critical radius and height, respectively, of a cylinder having the minimum volume for a given value of B^2 are

$$R = \frac{2.945}{B} \quad \text{and} \quad H = \frac{5.441}{B},$$

and in the present case, where B is 3.2×10^{-2} cm^{-1}, it follows that the required critical dimensions are radius 92 cm and height 170 cm.

The cross-sectional area of this cylinder is $(\pi)(92)^2 = 2.7 \times 10^4$ cm^2, and since the fuel rods are $5\frac{1}{2}$ in. apart, the area per unit cell is 30 in.^{2}, or 190 cm^2. There are thus $2.7 \times 10^4/190 = 140$ vertical fuel rods required for the critical reactor. The length of each rod is equal to H, i.e., 170 cm, and its radius, as given above, is 1.4 cm. Taking the density of uranium metal as 19 g/cm^3, it is readily found that the mass of fuel in the bare critical reactor is about 2.9×10^6 grams, or roughly 3.2 tons.

REFLECTED REACTORS

Effects of Reflector

3.201. The critical mass of a reactor can be decreased by surrounding the *core*, i.e., the region containing the fuel, with a blanket of a good scattering material, e.g., heavy water, graphite, or beryllium for a thermal reactor. This blanket acts as a *reflector*, since it scatters back into the chain-reacting system neutrons which would otherwise have been lost as a result of leakage. Since a reflector reduces neutron leakage from the reactor core, it permits a given fuel-moderator system to become critical when the dimensions are appreciably less than required for a bare reactor. The use of a reflector thus results in a saving of fissionable material.

3.202. In addition to decreasing the volume (and mass) of the core, the reflector can make possible an increase in the power output of the reactor for a given mass of fuel. As seen in § 2.163, the power level is proportional to the average thermal-neutron density or flux in the core. Suppose that in a reactor with reflector, the flux in the center of the core is the same as that for a bare reactor. Because of the return of neutrons to the core by the reflector, the flux near the boundaries will be much greater than in the absence of a reflector (Fig. 3.19). The average thermal-neutron flux over the whole core, and hence the power output under given conditions, will then be increased by the reflector. The multiplying properties of the outermost regions of the core are used more effectively when a reflector is employed.

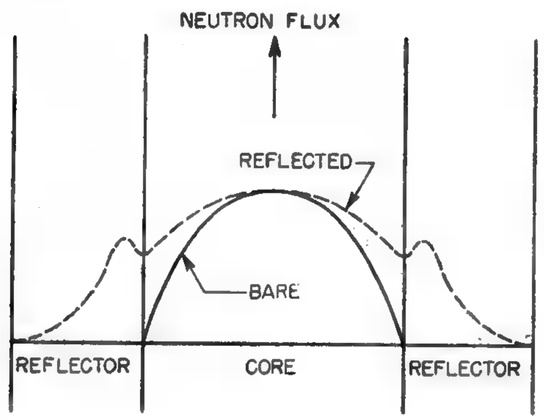


Fig. 3.19. Approximate thermal-neutron flux distributions in bare and reflected reactors

One-group Treatment

3.203. With the exception of reactors for which the slowing down properties of the reflector are identical with that of the core, the solution of the age-diffusion equation for a reflected reactor can, in general, be obtained only by numerical procedures. As an alternative, an approximate approach, known as the *group method*, has been developed and used with a fair degree of success. In this treatment, it is postulated that neutrons can be divided into a number of energy intervals or energy groups, from source energy down to thermal values. Within each group the neutrons are assumed to diffuse without energy loss, i.e., according to a monoenergetic diffusion equation, until they have suffered the average number of collisions which would be required to decrease their energy to the next (lower) group. At this point, it is supposed that the neutrons are suddenly transferred to the latter group. This process continues until the neutron energy is degraded from the group of highest (fission) energy to that of the lowest (thermal) energy.

3.204. The simplest approximation results from considering all the neutrons to be in a single group; in other words, it is supposed that all production, diffusion, and absorption of neutrons occur at a single energy, namely, the thermal energy. Although this represents a very severe approximation for an actual thermal reactor, it serves to illustrate some general principles. Preliminary results obtained very simply by the one-group method can be refined later by more involved procedures.

3.205. Using the subscripts c and r to indicate core and reflector, respectively, the steady-state (critical) diffusion equation, in the absence of a primary source, for the one group of core neutrons is

$$D_c\nabla^2\phi_c - \Sigma_{ac}\phi_c + k\Sigma_{ac}\phi_c = 0, \tag{3.205.1}$$

where D_c is the diffusion coefficient and Σ_{ac} is the macroscopic absorption cross section of the core material for thermal neutrons. The third term in equation (3.205.1), i.e., the source term, is accounted for by the fact that k neutrons are produced for every one absorbed. Upon dividing through by D_c and rearranging, the result is

$$\nabla^2\phi_c + \frac{(k-1)}{L_c^2}\phi_c = 0,$$

where L_c^2 for the core neutrons is equal to D_c/Σ_{ac}. This equation may be written in the familiar form

$$\nabla^2\phi_c + B^2\phi_c = 0, \tag{3.205.2}$$

so that the buckling of the core is given by

$$B^2 = \frac{k-1}{L_c^2}. \tag{3.205.3}$$

This may be regarded as the one-group critical equation.

3.206. If equation (3.205.3) is transposed to

$$\frac{k}{1 + L_c^2B^2} = 1,$$

it is seen to be similar to the critical equation (3.108.2), except that it contains no factor for the nonleakage probability during slowing down. This is, of course, to be expected, since the one-group treatment is based on the postulate that all neutrons have the same energy. However, although equation (3.205.3) defines the buckling of the core according to one-group theory, for a large reactor it is preferable to use, for purposes of calculation, the expression

$$B^2 = \frac{k - 1}{M_c^2}, \tag{3.206.1}$$

where $M_c^2 = L_c^2 + \tau_c$, thus introducing a correction for slowing down.

3.207. The reflector contains no fuel material, and so there is no source term in the diffusion equation for the reflector; the result is

$$D_r \nabla^2 \phi_r - \Sigma_{ar} \phi_r = 0, \tag{3.207.1}$$

where D_r and Σ_{ar} refer to the reflector. This expression may also be written as

$$\nabla^2 \phi_r - \kappa_r^2 \phi_r = 0, \tag{3.207.2}$$

where $\kappa_r^2 = \Sigma_{ar}/D_r$, so that κ_r is the reciprocal of the diffusion length of neutrons in the reflector (§ 3.33).

Infinite Plane Slab

3.208. In order to determine the flux distribution in the critical reactor, it is convenient to consider a simple case which can be reduced to a one-dimensional problem, namely, a slab of finite thickness but infinite extension. Consider such a slab of thickness H, with a reflector on each side of thickness T, which includes the extrapolation distance (Fig. 3.20). The origin of the coordinates is taken at the plane of symmetry of the slab. Since, in the present case, the Laplacian operator is simply d^2/dx^2, the core diffusion equation (3.205.2) becomes

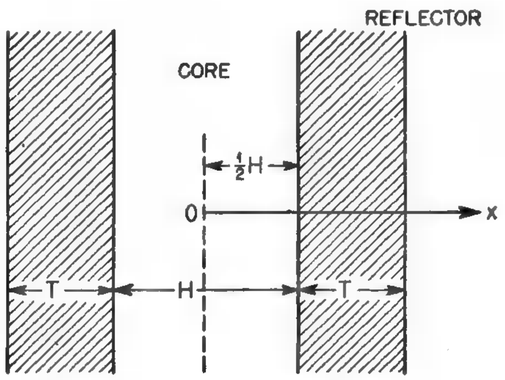


Fig. 3.20. Infinite slab reactor with reflector

$$\frac{d^2\phi_c(x)}{dx^2} + B^2\phi_c(x) = 0$$

of which the general solution, with B^2 positive, is

$$\phi_c(x) = A \cos Bx + C \sin Bx.$$

The requirements of symmetry rule out the sine term, and so the permissible solution for the infinite slab is

$$\phi_c(x) = A \cos Bx, \tag{3.208.1}$$

where A is an arbitrary constant.

3.209. The general solution of the reflector diffusion equation (3.207.2) involves hyperbolic sine and cosine terms, since κ^2 is positive; thus the neutron flux in the reflector is given by

$$\phi_r(x) = A' \cosh (\kappa_r x) + C' \sinh (\kappa_r x), \tag{3.209.1}$$

subject to the boundary condition that the flux is zero at the extrapolated boundary of the reflector, i.e., when $x = \frac{1}{2}H + T$. This leads to the result

$$\phi_r(\tfrac{1}{2}H + T) = A' \cosh \{\kappa_r(\tfrac{1}{2}H + T)\} + C' \sinh \{\kappa_r(\tfrac{1}{2}H + T)\} = 0,$$

so that

$$A' = -C' \tanh \{\kappa_r(\tfrac{1}{2}H + T)\}.$$

If this value for A' is now inserted into equation (3.209.1), the solution of equation (3.207.2) is

$$\phi_r(x) = C \sinh \{\kappa_r(\tfrac{1}{2}H + T - x)\}, \tag{3.209.2}$$

where C is a new arbitrary constant.

3.210. The arbitrary constants A and C may be related by introducing the boundary conditions that the neutron flux and current density shall be continuous at the core-reflector interface (see § 3.20), i.e., when $x = \frac{1}{2}H$; these conditions may be expressed as

$$\phi_c(\tfrac{1}{2}H) = \phi_r(\tfrac{1}{2}H)$$

and

$$D_c \frac{d\phi_c(\frac{1}{2}H)}{dx} = D_r \frac{d\phi_r(\frac{1}{2}H)}{dx}.$$

Using the first of these conditions, it follows from equations (3.208.1) and (3.209.2) that

$$A \cos (\tfrac{1}{2}BH) = C \sinh (\kappa_r T);$$

and from the second,

$$AD_cB \sin (\tfrac{1}{2}BH) = CD_r\kappa_r \cosh (\kappa_r T).$$

Upon dividing the second of these equations by the first, it is seen that

$$D_cB \tan (\tfrac{1}{2}BH) = D_r\kappa_r \coth (\kappa_r T). \tag{3.210.1}$$

This transcendental equation is the critical equation for the reflected infinite slab reactor, according to the one-group treatment.

3.211. The value of the buckling B^2 can be derived from the corrected equation (3.206.1), and $D_c (= \Sigma_{ac} L_c^2)$ can be obtained from the known properties of the core components; D_r is the diffusion coefficient of the reflector material and κ_r is the reciprocal of its thermal diffusion length. With these data available,

it is possible to calculate, from equation (3.210.1), the relationship between the thickness T of the reflector and the critical thickness H of the infinite slab core. The results of a typical case are shown by curve A in Fig. 3.21. It is seen that, as the reflector thickness is increased, the critical core thickness decreases rapidly at first, but beyond a certain thickness of reflector, the core size does not decrease appreciably. There is thus little to be gained by increasing the reflector thickness beyond this amount.

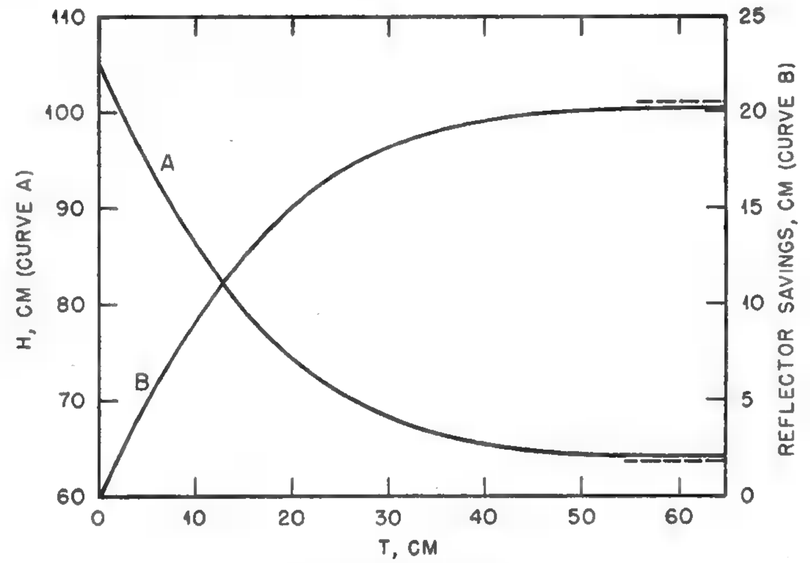


FIG. 3.21. Reflector savings for infinite slab reactor

REFLECTOR SAVINGS

3.212. The decrease in the critical size of a reactor, due to a reflector, is expressed by means of the *reflector savings*, δ, defined, in the case of an infinite slab, by

$$\delta \equiv \tfrac{1}{2}H_0 - \tfrac{1}{2}H, \tag{3.212.1}$$

where H_0 is the critical thickness of the bare reactor and H is that with a reflector. For the bare reactor, H_0 is equal to π/B (cf. Table 3.7),* and so

$$\delta = \frac{\pi}{2B} - \frac{H}{2}$$

or

$$\frac{H}{2} = \frac{\pi}{2B} - \delta.$$

Substituting this value for $\frac{1}{2}H$ into the critical equation (3.210.1), the result is

* It should be noted that the infinite slab is a rectangular parallelepiped in which two of the linear dimensions are infinite. The results obtained below can be applied to each of the three dimensions of a finite rectangular parallelepiped.

$$D_c B \tan\left(\tfrac{1}{2}\pi - B\delta\right) = D_r\kappa_r \coth\left(\kappa_r T\right)$$

or

$$D_c B \cot B\delta = D_r\kappa_r \coth\left(\kappa_r T\right).$$

Upon inverting and rearranging, this becomes

$$\tan B\delta = \frac{D_c B}{D_r\kappa_r} \tanh\left(\kappa_r T\right)$$

or

$$\delta = \frac{1}{B}\tan^{-1}\left\{\frac{D_c B}{D_r\kappa_r}\tanh\left(\kappa_r T\right)\right\}. \tag{3.212.2}$$

From this equation the reflector savings can be calculated for various specified thicknesses T of the reflector. The general nature of the variation of the reflector savings with the thickness of the reflector is shown by curve B in Fig. 3.21.

3.213. For a large reactor, B is small and so also is $B\delta$; then, after rearranging equation (3.212.2), $\tan B\delta$ may be replaced by $B\delta$, with the result that

$$\delta \approx \frac{D_c}{D_r\kappa_r}\tanh\left(\kappa_r T\right)$$

$$\approx \frac{D_c}{D_r}L_r \tanh\frac{T}{L_r}, \tag{3.213.1}$$

replacing κ_r by $1/L_r$, where L_r is the diffusion length in the reflector. If, in addition, the reflector is thick, so that T/L_r is large, $\tanh T/L_r$ approaches unity. The reflector savings is then given by equation (3.213.1) as

$$\delta \approx \frac{D_c}{D_r}L_r, \tag{3.213.2}$$

so that δ attains a limiting value with increasing reflector thickness, as is apparent from curve B in Fig. 3.21.*

3.214. When T/L_r is 2, i.e., when the thickness of the reflector is twice the diffusion length, $\tanh T/L_r$ is already 0.96, and the reflector savings will be 96 per cent of the limiting value given by equation (3.213.2). Thus, for the reasons given in § 3.44, a reflector having a thickness of about two thermal diffusion lengths behaves virtually as a reflector of infinite thickness, according to one-group theory. More exact calculations, which take into account the fact that the neutrons do not all have the same energy, show that a reflector thickness of 1.5 to 2 migration lengths (Table 3.5) is almost equivalent to an infinite reflector, as far as the effect on the critical mass is concerned.

Example Estimate the amount of fuel required for a natural uranium-graphite reactor, in the form of a cube, which is surrounded on all sides by a graphite reflector 100 cm thick.

The migration length of thermal neutrons in graphite is about 54 cm, and so the reflector may be regarded as almost equivalent to one of infinite thickness. Hence,

* Equations (3.213.1) and (3.213.2) are also applicable to a large spherical reactor, where δ is the decrease in the radius due to a reflector of thickness T.

treating each dimension of the cubic reactor as the finite dimension (H) of an infinite slab, the reflector savings is given by equation (3.213.2). Since the moderator and reflector are of the same material, D_c/D_r will not be very different from unity and the reflector savings will be equal to the diffusion length in the reflector, i.e., 50 cm in graphite. By equation (3.212.1) the reflector savings is the decrease in the half-thickness of the slab, and so the total savings in the length of the cube edge will be about 100 cm.

In the worked example dealing with the bare reactor, the edge length of the critical reactor was found to be 560 cm. Consequently for the reflected reactor under consideration the edge length of the core will be 460 cm. The volume of the reactor core is thus $(460/560)^3$, i.e., 0.55, of that of the bare reactor. Assuming the same radius of fuel rods and the same lattice spacing, the mass of fuel is $54 \times 0.55 = 30$ tons, instead of 54 tons in the bare reactor. (It may be mentioned that this represents the amount of fuel required to make the reactor just critical. For various reasons, to be considered in Chapter VI, additional fuel is always included when the reactor is constructed.)

3.215. An examination of equations (3.212.2), *et seq.*, shows that the effectiveness of a particular reflector material is determined to a great extent by the ratio L_r/D_r; because L_r^2 is equal to D_r/Σ_{ar}, it is the value of $1/\Sigma_{ar}D_r$ (or its square root) which is important. Since D_r is proportional to the transport mean free path (§ 3.24), and so is inversely related to the transport (and scattering) cross sections, it follows that Σ_{sr}/Σ_{ar} is the property of the reflector which is important. In other words, a good reflector will be a material having a high scattering cross section and a low absorption cross section. This general conclusion is in agreement with the results of a qualitative consideration of the properties to be expected from a good reflector.

3.216. There is another desirable property of a reflector which is not apparent from one-group theory. An appreciable proportion of the neutrons leaking from a reactor core do so during the slowing down process. These will be neutrons with energies in excess of the thermal value. It would be advantageous if they were returned to the core with their energies reduced; this can be achieved by using a good thermalizing material, i.e., an element having a large ξ, as reflector. As a rough generalization, therefore, it may be stated that for a good (thermal) reflector $\xi\Sigma_s/\Sigma_a$, i.e., the moderating ratio (§ 3.63), should be high. From the values given in Table 3.3, it is seen that heavy water would be an excellent reflector material for a thermal reactor.

Ratio of Maximum to Average Flux in Slab Reactor

3.217. The critical neutron flux distribution, in the x-direction, for an infinite-slab reflected reactor is given by equation (3.208.1). The maximum flux, which is that along the plane of symmetry, i.e., when x is zero, is then equal to the arbitrary constant A; this is determined by the power level of the reactor. The average flux is obtained by integrating $\phi_c(x)$ over the thickness of the slab and dividing by the thickness; thus,

$$\phi_{av} = \frac{1}{H}\int_{-\frac{1}{2}H}^{\frac{1}{2}H} A \cos Bx\, dx$$

$$= \frac{2A}{BH}\sin(\tfrac{1}{2}BH).$$

Since the maximum flux ϕ_{max} is equal to A, the ratio of maximum to average flux in the critical reactor is

$$\frac{\phi_{max}}{\phi_{av}} = \frac{\frac{1}{2}BH}{\sin(\frac{1}{2}BH)}. \tag{3.217.1}$$

3.218. This expression combined with the critical equation (3.210.1) may be used to calculate the flux ratio for various thicknesses of core and reflector. Particular values for H and T are chosen, and then, with D_c, D_r, and κ_r known, the equation (3.210.1) is solved for B by a process of iteration. With B and H known for a given value of T, the ratio expressed by equation (3.217.1) can be calculated. For zero thickness of reflector, i.e., for a bare reactor, B is equal to π/H, and hence the ratio ϕ_{max}/ϕ_{av} is $\frac{1}{2}\pi$, i.e., 1.57. At the other extreme, for large reflector thicknesses, i.e., as $T \to \infty$, $\coth(\kappa_r T) \to 1$, and from equation (3.210.1), $\frac{1}{2}BH = \tan^{-1}(D_r\kappa_r/D_cB)$. The limiting value of ϕ_{max}/ϕ_{av} can then be obtained for any core thickness.

3.219. The results for a reactor having beryllium as both moderator and reflector are given in Fig. 3.22 for infinite slabs of 25, 50, and 75 cm thickness, with reflector thicknesses (including extrapolation distances) up to 50 cm. The

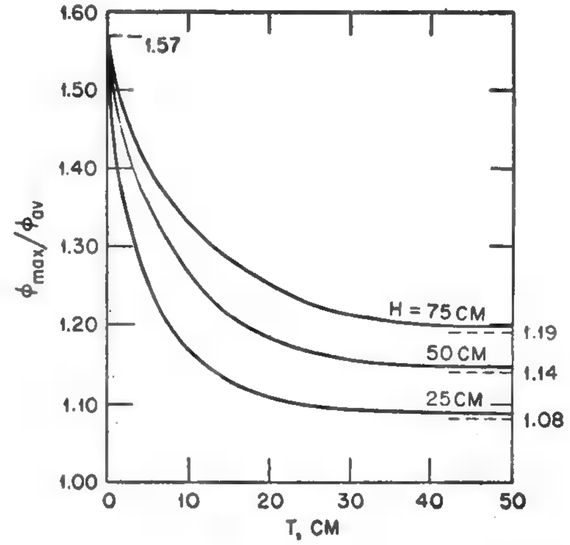


Fig. 3.22. Ratio of maximum to average flux as function of reflector thickness

initial sharp decrease in the ratio of maximum to average flux with increase in reactor thickness indicates a marked flattening of the flux distribution in the reactor core resulting from the presence of the reflector. With increasing thickness of the latter, however, a limiting value of the flux ratio is approached asymptotically when the reflector thickness is roughly twice the diffusion length

of thermal neutrons in the reflector, i.e., about 48 cm in this case. Further increase in thickness has little effect on the flux distribution. This conclusion is in general agreement with the results obtained above with regard to the effect of the reflector on the critical size.

Two (or More) Groups of Neutrons

3.220. The one-group treatment is highly approximate, but it is very simple and leads to conclusions which are qualitatively correct. However, the reflector savings calculated on the basis of one group of neutrons are too low for at least two reasons. In the first place, since the one-group method assumes all neutrons to be thermal, no allowance is made for the fact that fast neutrons escaping from the core have a longer average life in the reflector than do thermal neutrons and so they have a higher probability of being scattered back into the core. Further, neutrons entering the reflector with energies above the resonance level are moderated in the reflector; they then return to the core as thermal neutrons, having completely avoided the resonance capture to which they would have been liable if they had reached thermal energies in the core. Both these factors tend to increase the effectiveness of the reflector, as compared with that to be expected if all the neutrons had thermal energies.*

3.221. A second approximation to the treatment of reflected reactors is to use the two-group theory. It is then postulated that all the neutrons can be divided into two groups, namely, fast (fission) and slow (thermal) neutrons. There are now four differential equations; two are similar to equation (3.205.1), one for each neutron group in the core, and two others, like equation (3.207.1), give the fast and slow flux distributions in the reflector. In solving these equations, six boundary conditions are used: continuity of both fast- and slow-neutron flux and neutron current density at the core-reflector boundary, and zero fast- and slow-neutron flux at the (same) extrapolated boundary of the reflector. The actual solution is somewhat involved and will not be given here.

3.222. The group method has been refined still further by introducing additional neutron-energy groups. The solution of the differential equations obtained is a major problem, but several methods have been described, e.g., solution in closed form, series expansion, method of iteration, or a combination of these procedures.

3.223. There is one result of the two- (and higher-) group treatment which is worth mentioning; it refers to the spatial distribution of the neutron flux. For convenience, and in accordance with the two-group theory, the neutrons are divided into two groups; thermal neutrons and all other (fast) neutrons, respectively. The general nature of the variation of the flux with distance from the center of the reactor core is shown in Fig. 3.23. It will be observed that the thermal-neutron flux exhibits a maximum within the reflector at a short distance

* A reasonably good empirical rule is that the reflector savings is 1.2 times that calculated by the one-group method.

from the core-reflector interface. This may be ascribed to the fact that thermal neutrons are produced in the reflector by the slowing down of fast ones, but they are absorbed very much less strongly in the reflector than in the core. Farther from the interface the thermal flux, like the fast-neutron flux, falls to zero at the extrapolated boundary of the reflector. It is because the thermal-neutron flux within a good reflector may be as high as, or higher than, in the core that some

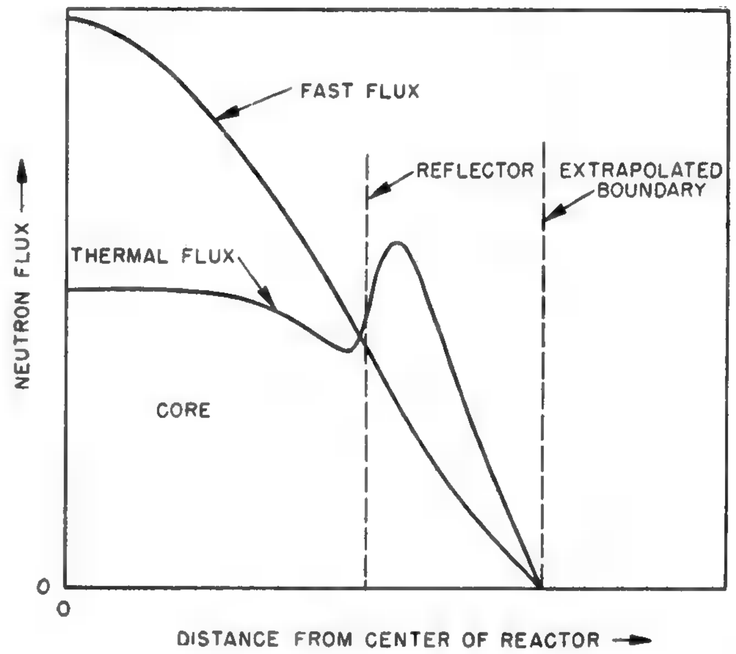


Fig. 3.23. Thermal and fast neutron flux distributions in reflected reactor

thermal reactors can be controlled by means of an absorber inserted into the reflector near the core, instead of into the core itself.

INTERMEDIATE AND FAST REACTORS

The Intermediate Reactor*

3.224. An intermediate reactor is one which operates mainly on fissions caused by the absorption of neutrons with energies in the intermediate range, e.g., up to about 1000 ev. In general, intermediate reactors are characterized (1) by smaller nonleakage probabilities for neutrons while slowing down than in thermal reactors, (2) by having cores containing relatively large amounts of nonfission absorbers of thermal neutrons, i.e., thermal poisons, or (3) by a combination of both these circumstances.

3.225. In the first case, either the reactor may be smaller than a thermal reactor or the moderating efficiency of the core material may be less than for a

* For the possible advantages of intermediate reactors, see § 1.121.

thermal reactor of the same size. This latter condition would arise in a reactor in which the core consisted partly of moderator, e.g., carbon or beryllium, and partly of a poorly moderating coolant, e.g., liquid sodium, with a relatively small slowing down power. As a result of the partial replacement of good moderator by the coolant, which is a poor moderator, the leakage of neutrons during the slowing down process is increased.

3.226. In order to make the system critical, the infinite multiplication factor k must be larger in this case than it would need to be if the entire core volume were occupied by moderator and fuel only. The increase in k is achieved by increasing the ratio of fuel to moderator; as a result, there is an increase in the probability that neutrons will be absorbed in the intermediate region before they become thermal. In an enriched fuel, this will mean a larger proportion of fissions will be due to intermediate-energy neutrons than is the case in thermal reactors.

3.227. If the reactor core contains a relatively large proportion of thermal poisons, in the form of structural material, cladding for fuel elements, etc., additional fuel must be used to compensate for the increased loss of neutrons by parasitic capture. For the reasons given in the preceding paragraph, this may result in an appreciable increase in the absorption by the fuel of, and hence fission production by, neutrons in the intermediate energy range.

3.228. The theoretical treatment of intermediate reactors is much more involved than is that of thermal reactors, as described in the earlier parts of this chapter. In a thermal reactor essentially all the fissions are caused by thermal neutrons, and reasonably good results are obtained by the use of the diffusion equation for monoenergetic neutrons. In an intermediate reactor, however, the neutrons causing fission cover a considerable energy range. The most satisfactory treatment of such systems developed so far is the group method, the neutrons being divided into ten or more energy groups, ranging from fission to thermal energies. An age-diffusion type of equation is then applied to each neutron group. Since the solution of the problem is complicated, it will not be given here.*

The Fast Reactor

3.229. In a reactor containing no moderator, and in which the coolant and structural material are made up of elements of fairly high mass numbers (or atomic weights) so that they are poor moderators, the fissions will be caused almost entirely by neutrons of high energy. Such a system would constitute a fast reactor. There may be some slowing down of neutrons due to inelastic collisions but, since such collisions are usually significant only for energies in excess of about 0.1 Mev (§ 2.100), it is evident that there will be very few neutrons with energies less than this value. The average neutron energy in a fast reactor may be taken as about 0.2 Mev, and so the fuel must contain a large

* See H. Hurwitz, Report AECD-3019; R. Ehrlich and H. Hurwitz, Jr., *Nucleonics*, **12**, No. 2, 23 (1954); R. B. Spooner, *ibid.*, **12**, No. 4, 36 (1954).

proportion of uranium-235 or plutonium-239, although a certain amount of other high atomic weight (nonmoderating) material may be present.

3.230. In a fast reactor, the infinite multiplication factor is essentially equal to η, the average number of fast neutrons produced for each neutron absorbed in fuel. This means that k may be greater than 2 (see Table 1.7); the critical reactor will thus be small, since considerable neutron leakage is permissible. Because of the small size, the conditions for applicability of the diffusion theory approximation are not valid, for there is probably no point in the reactor that is more than two scattering (or transport) mean free paths from a boundary.

3.231. In the calculation of the critical dimensions of a fast reactor it is consequently necessary to use transport theory. Because of the relatively limited range of the neutron energies, the neutrons may be treated as monoenergetic, in first approximation. The equation which relates the properties of the multiplying system to the buckling, B^2, is found to be

$$\frac{\nu\Sigma_f}{B}\tan^{-1}\left(\frac{B}{\Sigma}\right) \approx 1,$$

where Σ_f and Σ are the macroscopic fission and total cross sections of the medium, respectively, for fast neutrons. From this expression an estimate can be made of the dimensions of the critical fast reactor, including the extrapolation distance. In order to obtain the actual critical dimensions, the extrapolation distance must be known, especially as it constitutes an appreciable proportion of the total. It is not generally equal to $0.71\lambda_t$ as given in § 3.25, and its exact value can be obtained only by complicated methods.

Symbols Used in Chapter III

A	atomic weight (or mass number)
A, A'	arbitrary constants
a	resonance absorption probability
a	thickness of diffusion medium
B^2	buckling
C	center of mass system
C, C'	arbitrary constants
cosh	hyperbolic cosine; $\cosh x = \frac{1}{2}(e^x + e^{-x})$
coth	hyperbolic cotangent (1/tanh)
D	diffusion coefficient (of neutron flux)
D_0	diffusion coefficient (of neutron density)
D_c, D_r	diffusion coefficient in core, reflector
d	extrapolation distance
E	neutron energy
E_0	initial (fission) neutron energy
E_1	neutron energy before collision
E_2	neutron energy after collision

E_{th}	thermal neutron energy
$F(E)$	collision density, i.e., collisions/(cm³)(sec), for neutrons of energy E per unit energy interval
f	thermal utilization
f_r	resonance utilization
H	height of cylinder
H	thickness of reflected slab reactor
H_0	thickness of bare slab reactor
J_0	Bessel function of first kind and zero order
$\mathbf{J}$	neutron current vector, neutrons/(cm²)(sec)
J_x	neutron current in x-direction
J_+, J_-	neutron currents in opposite directions
k	infinite multiplication factor
k_{eff}	effective multiplication factor
L	laboratory system
L	diffusion length
L_c, L_r	diffusion length in core, reflector
L_m	diffusion length of moderator
l	thermal neutron lifetime in finite medium
l_0	thermal neutron lifetime in infinite medium
l_m	thermal neutron lifetime in moderator
M	mass (of fuel lump)
M	migration length
M^2	migration area
m	moderator
m	neutron mass
N	atoms (or nuclei)/cm³
N_a	Avogadro number
N_m	moderator atoms (or nuclei)/cm³
N_u	fuel atoms (or nuclei)/cm³
n	neutron density, neutrons/cm³
n	integer
p, $p(E)$	resonance escape probability (at energy E)
Q	neutron source strength
q	slowing down density, neutrons/(cm³)(sec)
q^*	slowing down density in absorbing medium
$q(E)$	slowing down density at energy E
$q(\tau)$	slowing down density at age τ
$q(\tau, \mathbf{r})$	slowing down density at age τ and position represented by coordinate $\mathbf{r}$
R	radius of sphere or cylinder
$R(r)$	space function of neutron age
$\mathbf{r}$	general spatial coordinate

r	radial distance, polar coordinate
$\overline{r^2}$	mean square distance traveled by thermal neutron
$\overline{r_s^2}$	mean square distance traveled by neutron while slowing down
S	source term in diffusion equation
S	surface area of fuel lump
sinh	hyperbolic sine; $\sinh x = \frac{1}{2}(e^x - e^{-x})$
T	thickness of reflector
$T(t)$	time function of neutron age
t	time
tanh	hyperbolic tangent ($\tanh x = \sinh x/\cosh x$)
u	fuel
V	volume
V_i	volume of impurity, poison, etc.
V_m, V_u	volume of moderator, fuel
v	neutron velocity
x, y, z	coordinates
z	ratio of fuel/moderator macroscopic absorption cross sections (Σ_{au}/Σ_{am})
α	ratio of capture to fission cross sections
α	$(A-1)^2/(A+1)^2$
$-\gamma$	slope of plot in exponential experiment
δ	reflector savings
ϵ	fast-fission factor
η	fission neutrons per neutron absorbed in fuel (average)
θ	polar coordinate
θ	scattering angle in C system
κ	Σ_a/D, reciprocal of diffusion length
κ_r	value of κ in reflector
λ_a	absorption mean free path
λ_s	scattering mean free path
λ_t	transport mean free path
$\bar{\mu}_0$	average cosine of neutron scattering angle
ν	fission neutrons per fission (average)
ξ	average logarithmic energy decrement per collision
ρ	density
Σ	total macroscopic cross section
Σ_a	macroscopic absorption cross section
Σ_{ac}, Σ_{ar}	absorption cross section of core, reflector
Σ_{am}, Σ_{au}	absorption cross section of moderator, fuel
Σ_s, $\Sigma_s(E)$	macroscopic scattering cross section (at energy E)
Σ_{sm}, Σ_{su}	scattering cross section of moderator, fuel
σ_a	microscopic absorption cross section
σ_{am}, σ_{au}	absorption cross section of moderator, fuel

σ_s	microscopic scattering cross section
σ_{sm}, σ_{su}	scattering cross section of moderator, fuel
τ	Fermi age, cm^2
$\tau(E)$	Fermi age of neutrons of energy E
τ_m	Fermi age in moderator
τ_{th}	Fermi age of thermal neutrons
ϕ	neutron flux, neutrons/(cm^2)(sec)
ϕ_{av}, ϕ_{max}	average flux, maximum flux in reactor core
$\phi(E)$	neutron flux at energy E per unit energy interval
$\phi(\mathbf{r})$	neutron flux at point having coordinate $\mathbf{r}$
$\phi(x)$	neutron flux at point having coordinate x
ϕ_m, ϕ_u	neutron flux in moderator, fuel
ϕ_c, ϕ_r	neutron flux in core, reflector
φ	polar coordinate
Ψ	scattering angle in L system
Ω	solid angle
∇^2	Laplacian operator

PROBLEMS

1. Calculate (*a*) the diffusion coefficient, (*b*) the extrapolation distance, and (*c*) the diffusion length, of thermal neutrons in graphite from the scattering and absorption cross sections (4.8 barns and 0.0045 barn, respectively). The density of graphite is to be taken as 1.62 g/cm^3.

2. A point source, emitting Q neutrons/sec, is located at the center of a sphere, finite radius R, including the extrapolation distance, of a nonmultiplying medium bounded by a vacuum. Derive an expression for the spatial distribution of the neutron flux in the sphere, as a function of the radial distance r from the center.

3. From the expression derived in the preceding problem, plot the variation of Q/ϕ as a function of distance from the source for spheres of radii (*a*) twice the diffusion length, (*b*) three times the diffusion length, (*c*) infinity. What conclusions can be drawn from the results?

4. By treating an infinite plane source of monoenergetic neutrons as made up of an infinite number of point sources spread over the plane, use equation (3.33.1) to show that the flux distribution in the x-direction perpendicular to the plane source, emitting Q neutrons/(cm^2)(sec) in an infinite, nonabsorbing medium, is represented by $Qe^{-\kappa x}/2\kappa D$.

5. Show that the average net vector distance, $\bar{r}$, traveled by a thermal neutron, from the point at which it just becomes thermal to that at which it is captured, is equal to twice the diffusion length. Compare the average net vector (crow-flight) distance traveled by the thermal neutron with the corresponding total scalar (zigzag) distance in graphite. (Use the data in Problem 1.)

6. Recalling that commercial heavy water consists of 99.75 per cent D_2O and 0.25 per cent H_2O, use the following data to calculate the slowing down power and moderating ratio of heavy water. Scattering cross sections for epithermal neutrons: H, 23 barns; D, 3.3 barns; O, 4.0 barns. Absorption cross sections for thermal neutrons: H, 0.33 barn; D, 0.00046 barn; O, negligible.

7. The diffusion length of thermal neutrons in heavy water is quoted as 100 cm. From the data in the preceding problem estimate the diffusion length in pure D_2O. Consider the advantages and disadvantages of the effect of the 0.25 per cent of H_2O on the value of heavy water as a component of thermal reactors.

8. Derive an expression relating the so-called slowing down length to the actual mean net vector distance, $\bar{r}_s$, traveled by a neutron while slowing down. Taking the average scattering cross section of graphite in the range from 2 Mev to 0.025 ev to be 4 barns, compare the two lengths for the slowing down of neutrons over this energy range.

9. Derive the expression $B^2 = (\pi/H)^2$ for the buckling of an infinite slab reactor having a finite thickness H, including the extrapolation distance, and show that the flux distribution in the critical system is given by $\phi(x) = A \cos(\pi x/H)$, where A is a constant and x is the distance from the central plane in the finite dimension of the slab.

10. In a bare spherical reactor of radius R compare (*a*) the average neutron flux and (*b*) the flux at a radial distance from the center equal to $0.5R$, with the flux at the center. At what radial distance from the center will the local flux be equal to the average flux?

11. Using the expression in Table 3.7 for the buckling of a finite cylinder, show that there is a limiting radius below which a cylindrical thermal reactor cannot become critical, for a given fuel-moderator ratio.

12. Determine the minimum mass ratio of plutonium-239 to water that will just sustain a chain reaction in an infinitely large system.

13. Calculate the critical volume at ordinary temperatures of a cubical thermal reactor containing heavy water as moderator and pure uranium-235 as fuel in the atomic (or molecular) ratio of 10^5 to 1, assuming the continuous slowing down model to be applicable. Estimate the critical mass of uranium-235 for this reactor.

14. In the preceding example, what fraction of the neutrons produced in fission escape (*a*) while slowing down, (*b*) while at thermal energies? Calculate the mean lifetime of thermal neutrons in the reactor and compare the value with that in the moderator (heavy water) alone.

15. Determine the size of a bare spherical reactor (Water Boiler) containing uranium-235 and ordinary water in the weight ratio of 1 to 15, using equation (3.162.1). Compare the result with that given by the use of the continuous slowing model.

16. Estimate the error in the preceding problem resulting from the neglect of the neutron capture by sulfur and oxygen if the uranium were present in solution as uranyl sulfate (UO_2SO_4).

17. What would be the approximate critical mass of uranium-235 for the reactor referred to in Problem 15 if the sphere were surrounded by a thick graphite reflector?

18. Determine the values of the infinite multiplication factor in homogeneous mixtures of heavy water and natural uranium in the atomic (or molecular) ratios of 100, 200, 300, and 400 to 1, respectively. Hence estimate the maximum value of this factor, if parasitic capture of neutrons is neglected.

19. A heterogeneous reactor is constructed of natural uranium rods, of 1 in. diameter, arranged in a square lattice with a pitch of 6 in. suspended in a cylindrical vessel containing heavy water as moderator. Estimate the number of fuel rods required to make the reactor critical and the atomic (or molecular) ratio of moderator to fuel in the reactor. Compare the value of the infinite multiplication factor for this composition in a homogeneous system with that for the given heterogeneous system.

20. Assuming that all neutrons captured in uranium-238, either while slowing down or when thermal, produce plutonium-239, derive an expression for the ratio of plutonium formed to uranium-235 consumed in a reactor, in terms of p, f, and the uranium cross sections. Discuss the design conditions which might be used in a production reactor to obtain a high yield of plutonium-239. (Assume that neutron leakage while slowing down is negligible.)

21. A cylindrical reactor is to be constructed with fuel rods of 1.2 per cent enriched uranium, i.e., containing 1.2 atomic per cent of uranium-235, and ordinary water as

moderator. The rods are to be 0.5 in. diameter and placed 2 in. apart. Using the "two-group" equation (3.123.2), make an approximate estimate of the size of the reactor and of the weight of uranium-235 (in pounds) which would be required for the enrichment of natural uranium to prepare the necessary fuel.

22. In the Oak Ridge natural uranium-graphite reactor the 4-in. long and 1.1-in. diameter fuel slugs, clad (including the ends) with a 0.035-in. coating of aluminum, lie in channels of 1.75-in. square cross section through which the cooling air flows. Estimate the error in the thermal utilization (and hence in the multiplication factor) resulting from the neglect of the neutron absorption in the aluminum and air in the example following § 3.177. (The air may be assumed to be roughly at atmospheric temperature and pressure and the neutron capture may be attributed to the nitrogen which constitutes 78 atomic per cent of the air.)

23. A homogeneous reactor contains uranium-235 and graphite in the proportion of 1 to 10^4. Assuming this to be in the form of an infinite slab, determine by the one-group method the critical thickness (H) for graphite reflector thicknesses (T) of (a) 25 cm, (b) 50 cm, (c) 75 cm, and (d) 100 cm. Plot H as a function of T and draw a general conclusion from the result.

Chapter IV *

REACTOR THEORY: DISTURBANCE OF THE STEADY STATE

REACTOR KINETICS

Prompt Neutrons

4.1. The preceding chapter has dealt with the theory of nuclear reactors in the steady state, or equilibrium state, i.e., when the neutron density is constant and independent of time. During start-up and shut-down, as well as a result of transient changes, e.g., of temperature, occurring within the reactor, the neutron density changes and a knowledge of the rate of change under various conditions is desirable. The information so obtained is used in the design of control mechanisms, as will be seen in Chapter VI. The theoretical treatment given below refers strictly to bare, homogeneous, thermal reactors, but the general conclusions are applicable to reactors of all types.†

4.2. The diffusion equation (3.17.2) for thermal neutrons in a nonequilibrium, i.e., noncritical, system can be written as

$$D\nabla^2\phi - \Sigma_a\phi + S = \frac{\partial n}{\partial t} = \frac{1}{v} \cdot \frac{\partial \phi}{\partial t}, \tag{4.2.1}$$

where v is the mean velocity of the thermal neutrons; the thermal flux is then equal to nv, with n being the density of thermal neutrons. It was seen in § 3.103 that, according to the continuous slowing down model, i.e., Fermi age theory, the thermal-neutron source term due to fissions is $pq(\tau_{\text{th}})$, and using equation (3.107.2) this is seen to be

$$S = k\Sigma_a\phi e^{-B^2\tau}, \tag{4.2.2}$$

where $k\Sigma_a\phi$ is the number of thermal neutrons per cm³ per sec that would result from fissions in an infinite system, i.e., if there were no leakage. The factor

* Reviewed by M. C. Edlund.

† For reflected reactor, see J. G. Bayly, *Canad. J. Phys.*, **31**, 182 (1953); H. Rumsey, Jr., *J. Appl. Phys.*, **25**, 1395 (1954).

$e^{-B^2\tau}$ represents the nonleakage probability during slowing down, where B^2 is the buckling (§ 3.120).*

4.3. Upon introducing equation (4.2.2) into (4.2.1), it is seen that in the absence of an extraneous (primary) source

$$D\nabla^2\phi + (ke^{-B^2\tau} - 1)\Sigma_a\phi = \frac{1}{v} \cdot \frac{\partial\phi}{\partial t}.$$

Dividing through by Σ_a and making use of the fact that D/Σ_a is equal to L^2, the square of the thermal diffusion length, the result is

$$L^2\nabla^2\phi + (ke^{-B^2\tau} - 1)\phi = l_0\frac{\partial\phi}{\partial t}, \tag{4.3.1}$$

where $1/v\Sigma_a$ has been replaced by l_0, the lifetime of the thermal neutrons in an infinite medium having the same composition as the reactor (§ 3.102).

4.4. In writing equation (4.2.2) for the thermal-neutron source term in the time-variable diffusion equation (4.2.1), the tacit assumption has been made that the neutrons produced in fission, apart from those leaking out while slowing down, are instantaneously available as thermal neutrons. Thus the slowing down time of the fission neutrons and the fact that the emission of some of these neutrons is delayed are neglected. It was seen in § 1.128 that the time rate of change of the neutron flux (or density) when the multiplication factor is altered is dependent on the generation time, i.e., on the average time between successive generations of thermal neutrons. This includes both the slowing down time and the diffusion time (or lifetime) of the thermal neutrons. Since the former is usually small in comparison with the latter (see Table 3.6), no serious error is involved if the slowing down time is ignored, at least in the consideration of thermal reactors.

4.5. The mean delay time of the delayed neutrons, on the other hand, is of the order of 0.1 sec and this is appreciably greater than the thermal-neutron lifetime. The neglect of the delay time is thus not justified. It is, nevertheless, instructive to proceed with the discussion on the basis of the assumption that all the fission neutrons are prompt. Subsequently, the treatment will be modified to take into account the effect of the delayed neutrons.

4.6. In order to solve equation (4.3.1), it will be assumed that the position (**r**) and time (t) variables can be separated by writing

$$\phi \equiv \phi(\mathbf{r}, t) = \phi(\mathbf{r})T(t), \tag{4.6.1}$$

where $\phi(\mathbf{r})$ is a function of the space coordinate only and $T(t)$ is a function of time only. Combination of equations (4.6.1) and (4.3.1) then gives

$$L^2\frac{\nabla^2\phi(\mathbf{r})}{\phi(\mathbf{r})} + ke^{-B^2\tau} - 1 = \frac{l_0}{T(t)} \cdot \frac{dT(t)}{dt}. \tag{4.6.2}$$

* The results obtained are independent of the slowing down model, and so the Fermi age treatment is used for convenience.

Suppose that at a certain time, represented by $t = 0$, the reactor has been made to undergo a sudden (or step) change in the multiplication factor, after which k remains constant. In these circumstances, the left side of equation (4.6.2) contains no time variable, and the right side no space variable.

4.7. If the system is not far from critical, the spatial distribution of the thermal flux can be represented, to a good approximation, by equation (3.108.1), i.e.,

$$\nabla^2\phi(\mathbf{r}) + B^2\phi(\mathbf{r}) = 0,$$

where B^2 is the buckling. Hence, substituting $-B^2\phi(\mathbf{r})$ for $\nabla^2\phi(\mathbf{r})$ in equation (4.6.2), and dividing through by $1 + L^2B^2$, the result is

$$\frac{ke^{-B^2\tau}}{1 + L^2B^2} - 1 = \frac{l_0}{1 + L^2B^2} \cdot \frac{1}{T(t)} \cdot \frac{dT(t)}{dt}. \tag{4.7.1}$$

The first term on the left side is the effective multiplication factor, k_{eff} (see § 3.125), and the first factor on the right is the mean lifetime l of thermal neutrons in a finite medium (§ 3.127); thus equation (4.7.1) can be reduced to

$$k_{\text{eff}} - 1 = \frac{l}{T(t)} \cdot \frac{dT(t)}{dt}. \tag{4.7.2}$$

4.8. The excess of k_{eff} over unity is the excess multiplication factor k_{ex}, defined in § 1.126, i.e., $k_{\text{ex}} = k_{\text{eff}} - 1$, and so equation (4.7.2) is equivalent to

$$k_{\text{ex}} = \frac{l}{T(t)} \cdot \frac{dT(t)}{dt}.$$

Integration of this expression gives

$$T(t) = Ae^{t(k_{\text{ex}}/l)} \tag{4.8.1}$$

so that, by equation (4.6.1),

$$\phi(\mathbf{r}, t) = A\phi(\mathbf{r})e^{t(k_{\text{ex}}/l)}. \tag{4.8.2}$$

At zero time, i.e., when $t = 0$,

$$\phi(\mathbf{r}, 0) = A\phi(\mathbf{r}),$$

and if this is represented by ϕ_0, it follows from equation (4.8.2) that

$$\phi(\mathbf{r}, t) = \phi_0e^{t(k_{\text{ex}}/l)}, \tag{4.8.3}$$

where ϕ_0 is the thermal-neutron flux in the reactor, at a point represented by the coordinate **r**, at the instant before the effective multiplication factor underwent a step change of magnitude k_{ex}. It will be noted that equation (4.8.3) is analogous to equation (1.126.3) for the time change of the neutron density derived in a more approximate manner.

4.9. The *reactor period*, T, or e-folding time, may be defined as the time required for the neutron flux (or density) to change by a factor e, so that

$$\phi(\mathbf{r}, t) = \phi_0e^{t/T}. \tag{4.9.1}$$

Consequently, in the present case, when all the neutrons emitted in the fission process are assumed to be prompt,

$$T = \frac{l}{k_{ex}}. \tag{4.9.2}$$

The thermal-neutron flux in a reactor in a nonequilibrium state, i.e., $k_{ex} \neq 0$, should then increase (or decrease) in an exponential manner, the rate depending on the ratio of the average lifetime of the thermal neutrons to the excess multiplication factor, as found in § 1.126.

Effect of Delayed Neutrons

4.10. A general discussion of the effect of delayed neutrons on the time rate of change of neutron density was given in Chapter I; the subject will now be considered in more detail. According to the arguments in § 3.105, the over-all rate of production of fission neutrons, including both prompt and delayed, is $(k/p)\Sigma_a\phi$ neutrons/(cm³)(sec). It was stated in § 2.167 that at least five groups of delayed fission neutrons have been identified; let β_i represent the fraction of the total number of fission neutrons which are delayed neutrons belonging to the ith group. The rate of formation in the fission process of nuclei which are the precursors of this group of delayed neutrons is thus $\beta_i(k/p)\Sigma_a\phi$ nuclei/(cm³)(sec). If C_i nuclei/cm³ is the concentration of these precursors, the normal rate of radioactive decay is $-\lambda_i C_i$ nuclei/(cm³)(sec), where λ_i sec^{-1} is the appropriate decay constant (§ 2.5). The net rate of formation of the delayed-neutron precursors of the ith group, representing the difference between the rate of production and the rate of decay, is consequently given by

$$\frac{\partial C_i}{\partial t} = \beta_i \frac{k}{p} \Sigma_a\phi - \lambda_i C_i. \tag{4.10.1}$$

It should be noted that C_i and ϕ are functions of both space (**r**) and time (t) coordinates but the arguments are omitted for simplicity of representation.

4.11. The thermal-neutron source term in the time-dependent diffusion equation (4.2.1) now consists of two parts: one for prompt and the other for delayed fission neutrons. The sum of the β_i values for all the groups of delayed neutrons is represented by β, and so $1 - \beta$ is the fraction of fission neutrons emitted promptly. Hence,

$$\text{Prompt-neutron source term} = (1 - \beta)k\Sigma_a\phi e^{-B^2\tau}, \tag{4.11.1}$$

which is the value given by equation (4.2.2) multiplied by $1 - \beta$.

4.12. The rate of formation of delayed neutrons in any group is numerically equal to the rate of decay of the precursor, i.e., to $\lambda_i C_i$, and hence,

$$\text{Total rate of formation of all the delayed neutrons} = \sum_{i=1}^{m} \lambda_i C_i \text{ neutrons/(cm}^3\text{)(sec)}, \tag{4.12.1}$$

the summation being taken over all the m groups of delayed neutrons. The thermal-neutron source term due to delayed neutrons is obtained upon multiplying this rate of formation by the nonleakage probability during the slowing down process and by the resonance escape probability.

4.13. Since the energies of the delayed neutrons are less than the average of the prompt neutrons (see Table 2.11), the nonleakage probability while slowing down will be somewhat larger for the delayed than for the prompt neutrons. However, the difference is small, and so the Fermi age of a thermal neutron may be assumed to be the same, irrespective of whether it originated as a prompt neutron or a delayed neutron. The nonleakage probability is therefore $e^{-B^2\tau}$, where τ is essentially the same as in Table 3.4, and if p is the resonance escape probability,

$$\text{Delayed-neutron source term} = pe^{-B^2\tau} \sum_{i=1}^{m} \lambda_i C_i. \tag{4.13.1}$$

Upon substituting the two contributions to the source term into the thermal diffusion equation (4.2.1) the result, in the absence of an extraneous source, is

$$D\nabla^2\phi - \Sigma_a\phi + (1 - \beta)k\Sigma_a\phi e^{-B^2\tau} + pe^{-B^2\tau} \sum_{i=1}^{m} \lambda_i C_i = \frac{1}{v} \cdot \frac{\partial\phi}{\partial t},$$

where ϕ and C_i are functions of both space and time.

4.14. It can be shown that, provided k remains constant, e.g., after a step change, the space and time variables of the flux are separable,* and then $\nabla^2\phi$ may be replaced by $-B^2\phi$, as in § 4.7, if the system is not far from critical. If the resulting equation is divided through by Σ_a, and L^2 substituted for D/Σ_a and l_0 for $1/\Sigma_a v$, as above, it is seen that

$$-(L^2B^2 + 1)\phi + (1 - \beta)ke^{-B^2\tau}\phi + \frac{pe^{-B^2\tau}}{\Sigma_a} \sum_{i=1}^{m} \lambda_i C_i = l_0 \frac{d\phi}{dt}.$$

This may be divided through by $1 + L^2B^2$, and the definitions of k_{eff} and l, given by equations (3.125.1) and (3.127.1), introduced; the result is

$$[k_{eff}(1 - \beta) - 1]\phi + \frac{pk_{eff}}{k\Sigma_a} \sum_{i=1}^{m} \lambda_i C_i = l \frac{d\phi}{dt}. \tag{4.14.1}$$

4.15. The problem now is to solve the differential equations (4.10.1) and (4.14.1). Since both are linear and first order, and since the space and time variables are separable, solutions of the form

$$\phi = \phi_0 e^{t\omega} \tag{4.15.1}$$

and

$$C_i = C_{i0} e^{t\omega} \tag{4.15.2}$$

* Since the concentration of each delayed-neutron precursor must be everywhere proportional to the flux, the C_i's are also separable into functions of space and time.

may be superimposed. It is then required to find the conditions on the parameters, represented by ω and having the dimensions of reciprocal time, which permit such solutions. It should be noted that ϕ_0 and C_{i0} are the values of the neutron flux and of the concentration of the precursor of the ith group of delayed neutrons, respectively, at time $t = 0$, at a specified point in the reactor. It will be recalled that $t = 0$ is the time at which the multiplication factor suffered a sudden change.

4.16. Upon substituting the expressions for ϕ and C_i into equation (4.10.1), it is found that

$$C_{i0}\omega e^{t\omega} = -\lambda_i C_{i0} e^{t\omega} + \beta_i \frac{k}{p} \Sigma_a \phi_0 e^{t\omega}$$

and hence

$$C_{i0} = \frac{k\Sigma_a\phi_0}{p} \cdot \frac{\beta_i}{\omega + \lambda_i}. \tag{4.16.1}$$

If the same expressions for ϕ and C_i are also introduced into equation (4.14.1), with C_{i0} given by equation (4.16.1), the result is

$$k_{\text{eff}}(1 - \beta) - 1 + k_{\text{eff}} \sum_{i=1}^{m} \frac{\lambda_i \beta_i}{\omega + \lambda_i} = l\omega. \tag{4.16.2}$$

The total fraction, β, of delayed neutrons is equal to the sum of the individual β_i's, and since $k_{\text{eff}} - 1$ is defined as k_{ex}, it follows that equation (4.16.2) may be written as

$$k_{\text{ex}} + k_{\text{eff}} \sum_{i=1}^{m} \left(\frac{\lambda_i \beta_i}{\omega + \lambda_i} - \beta_i \right) = l\omega$$

or

$$k_{\text{ex}} = l\omega + k_{\text{eff}} \sum_{i=1}^{m} \frac{\omega \beta_i}{\omega + \lambda_i}. \tag{4.16.3}$$

4.17. It is convenient, at this point, to introduce a quantity ρ, called the *reactivity*,* and defined as

$$\rho \equiv \frac{k_{\text{ex}}}{k_{\text{eff}}} = \frac{k_{\text{eff}} - 1}{k_{\text{eff}}}. \tag{4.17.1}$$

Hence, if equation (4.16.3) is divided through by k_{eff} it is seen that

$$\rho = \frac{k_{\text{ex}}}{k_{\text{eff}}} = \frac{l\omega}{k_{\text{eff}}} + \sum_{i=1}^{m} \frac{\omega \beta_i}{\omega + \lambda_i}. \tag{4.17.2}$$

Alternatively, since by equation (4.17.1) $k_{\text{eff}} = 1/(1 - \rho)$, it follows that

$$\rho = \frac{l\omega}{l\omega + 1} + \frac{1}{l\omega + 1} \sum_{i=1}^{m} \frac{\omega \beta_i}{\omega + \lambda_i}. \tag{4.17.3}$$

* Some writers use $\Delta k/k$ (or $\delta k/k$) as essentially equivalent to the precise definition of reactivity given here.

4.18. Equation (4.17.3) is the characteristic equation which relates the parameters ω to the nuclear properties of the materials present in the reactor, as represented by the quantities ρ, β_i, and λ_i. It is seen to be an algebraic equation of degree $m + 1$ in ω, so that there are $m + 1$ values of ω corresponding to each value of the reactivity ρ. Thus the variation of the flux with time, at a given point in the system, may be expressed by a linear combination of $m + 1$ terms of the form of equation (4.15.1), i.e.,

$$\phi = A_0 e^{t\omega_0} + A_1 e^{t\omega_1} + \cdots + A_i e^{t\omega_i} + \cdots + A_m e^{t\omega_m}, \tag{4.18.1}$$

where $\omega_0, \omega_1, \ldots, \omega_i, \ldots, \omega_m$ are the $m + 1$ roots of equation (4.17.3). The A_0, A_1, etc., are constants determined by the initial conditions of the reactor in the steady state, as will be seen below.

Stable Reactor Period: Positive Reactivity

4.19. If the reactivity ρ is positive, i.e., $k_{\text{eff}} > 1$, it is found that m roots are negative and one is positive; thus, in equation (4.18.1) ω_0 may be taken as positive, and ω_1, ω_2, etc., as negative. Numerically, each of the m negative values of ω is of the same order as one of the decay constants, λ_i, of the delayed-neutron precursors. As the time t, i.e., the time interval following the small step change in the reactivity, increases, the contributions of all terms beyond the first on the right of equation (4.18.1) decrease rapidly to zero, because of the negative ω values. In other words, these terms make a transient contribution to the neutron flux; they soon become negligible compared to the first term, which increases with time because of the positive ω value. Hence, after a very short time interval, equation (4.18.1) reduces to the first term only, i.e.,

$$\phi = A_0 e^{t\omega_0}. \tag{4.19.1}$$

Since ω_0 has the dimensions of reciprocal time, it may be set equal to $1/T$, so that

$$\phi = A_0 e^{t/T}.$$

4.20. It is evident, from the definition of the reactor period given in § 4.9, that T is here the reactor period after the lapse of sufficient time to permit the contributions of the transient terms to damp out. Consequently T, defined by

$$T \equiv \frac{1}{\omega_0}, \tag{4.20.1}$$

is called the *stable reactor period*. The quantities $1/\omega_1$, $1/\omega_2$, etc., are sometimes referred to as *transient periods*, but they are negative and have no physical significance as reactor periods in the sense that $1/\omega_0$ does. The ω_1, ω_2, etc., are merely values of the parameter ω which satisfy the characteristic equation of the problem considered here. They affect the rate at which the neutron flux changes for a short time only after the effective multiplication factor has been increased (or decreased).

Negative Reactivity

4.21. If ρ is negative, i.e., $k_{eff} < 1$, all $m + 1$ values of ω, which are solutions of equation (4.17.3) for a particular ρ, are negative. The expression for the flux as a function of time, as given by equation (4.18.1), will then consist of $m + 1$ terms with negative exponents. The magnitude of each term will thus decrease with time. The first term, with the smallest numerical (negative) value of ω_0, will decrease at a slower rate than the others, and will ultimately yield a stable (negative) period equal to $1/\omega_0$.

One Group of Delayed Neutrons

4.22. Some indication of the relative importance of the stable and transient terms in equation (4.18.1) may be obtained by considering the simple case in which there is assumed to be only one group of delayed neutrons with a decay constant, λ, equal to the properly weighted average of the five actual groups.* It will be seen later that this average is given by

$$\lambda = \beta / \sum_{i=1}^{m} \frac{\beta_i}{\lambda_i}. \tag{4.22.1}$$

The values of λ_i and β_i for the five groups of delayed neutrons from uranium-235, derived from the data in Table 2.11, are given in Table 4.1; it will be recalled from equation (2.7.1) that λ for any radioactive species is equal to $0.693/t_{\frac{1}{2}}$,

TABLE 4.1. DECAY CONSTANTS AND YIELDS OF DELAYED-NEUTRON PRECURSORS IN FISSION OF URANIUM-235

$t_{\frac{1}{2}}$ (sec)	λ_i (sec^{-1})	β_i	β_i/λ_i
0.43	1.61	0.00085	0.0005
1.52	0.455	0.00241	0.0053
4.51	0.154	0.00213	0.0138
22.0	0.0315	0.00166	0.0526
55.6	0.0125	0.00025	0.0200
	Total	0.0073	0.092

where $t_{\frac{1}{2}}$ is the half life. The average decay constant, as defined by equation (4.22.1), is thus $0.0073/0.092 = 0.080$ sec^{-1}.

4.23. For a single group of delayed neutrons, equation (4.17.2) becomes

$$\rho = \frac{l\omega}{k_{eff}} + \frac{\omega\beta}{\omega + \lambda}, \tag{4.23.1}$$

which is a quadratic in ω. Since it has been postulated that the system is not far from critical, k_{eff} is not greatly different from unity, so that equation (4.23.1)

* For a method of complete solution, taking into account all the delayed-neutron groups, using the Laplace transform, see H. S. Isbin and J. W. Gorman, *Nucleonics*, **10**, No. 11, 68 (1952). Alternatively, an electrical simulator may be employed (§ 4.56, *et seq.*).

can be reduced to

$$\rho \approx l\omega + \frac{\omega\beta}{\omega + \lambda}, \tag{4.23.2}$$

which can be readily solved for ω. Provided $(\beta - \rho + l\lambda)^2 \gg 2|l\lambda\rho|$, as will be the case for thermal reactors with positive reactivities of 0.0025 or less, and for all negative reactivities, i.e., for $\rho \ll \beta$, the two solutions of this equation are

$$\omega_0 \approx \frac{\lambda\rho}{\beta - \rho} \quad \text{and} \quad \omega_1 \approx -\frac{(\beta - \rho)}{l}. \tag{4.23.3}$$

Since these solutions hold provided ρ is less than β, it is evident that ω_0 represents the positive solution, the reciprocal of which is the stable reactor period.

4.24. For a single (average) group of delayed neutrons, the variation of the thermal flux with time is given by equation (4.18.1) as

$$\phi = A_0 e^{t\omega_0} + A_1 e^{t\omega_1}, \tag{4.24.1}$$

and the corresponding expression for the delayed neutron precursor is

$$C = B_0 e^{t\omega_0} + B_1 e^{t\omega_1}. \tag{4.24.2}$$

The problem of immediate interest is to determine the constants A_0 and A_1. This is achieved by writing equation (4.10.1) for the single precursor, i.e.,

$$\frac{\partial C}{\partial t} = -\lambda C + \beta \frac{k}{p} \Sigma_a \phi, \tag{4.24.3}$$

and substituting equations (4.24.1) and (4.24.2) for ϕ and C, respectively. Another expression for $\partial C/\partial t$ can be obtained by differentiating equation (4.24.2), and, upon comparing the coefficients, expressions are obtained relating A_0 to B_0 and A_1 to B_1.

4.25. If ϕ_0 is the steady-state flux of the reactor before the step change in the reactivity, i.e., at $t = 0$, it follows from equation (4.24.1) that it is equal to $A_0 + A_1$. Similarly, the steady-state value of the precursor concentration, C_0, is equal to $B_0 + B_1$, by equation (4.24.2). Further, in the steady state $\partial C/\partial t$ is zero, and so a relationship between C_0 and ϕ_0 can be derived from equation (4.24.3). With the information now available, and using equation (4.23.3) for ω_0 and ω_1, it is found, after neglecting certain terms of small magnitude, that

$$A_0 \approx \frac{\beta}{\beta - \rho}\phi_0 \quad \text{and} \quad A_1 \approx -\frac{\rho}{\beta - \rho}\phi_0. \tag{4.25.1}$$

The variation of the thermal flux with time is consequently given by equations (4.23.3), (4.24.1), and (4.25.1) as

$$\phi = \phi_0 \left[\frac{\beta}{\beta - \rho} e^{\frac{\lambda\rho t}{\beta - \rho}} - \frac{\rho}{\beta - \rho} e^{-\frac{(\beta - \rho)t}{l}} \right]. \tag{4.25.2}$$

If the reactivity is positive, then $\beta - \rho$ will be positive, under the conditions

postulated in the foregoing derivation, and so the first term in the brackets in equation (4.25.2) has a positive coefficient and the second a negative coefficient. The thermal-neutron flux is thus the difference of two terms, of which one (the first) increases while the other decreases with increasing time after the step change ρ in the reactivity.*

4.26. The second (or transient) term, with the negative exponent, dies out in a very short time. Subsequently the flux variation is given by the first term of equation (4.25.2), namely,

$$\phi \approx \phi_0 \frac{\beta}{\beta - \rho} e^{\frac{\lambda \rho t}{\beta - \rho}}. \tag{4.26.1}$$

so that the stable reactor period, assuming one (average) group of delayed neutrons, is

$$T \approx \frac{\beta - \rho}{\lambda \rho}. \tag{4.26.2}$$

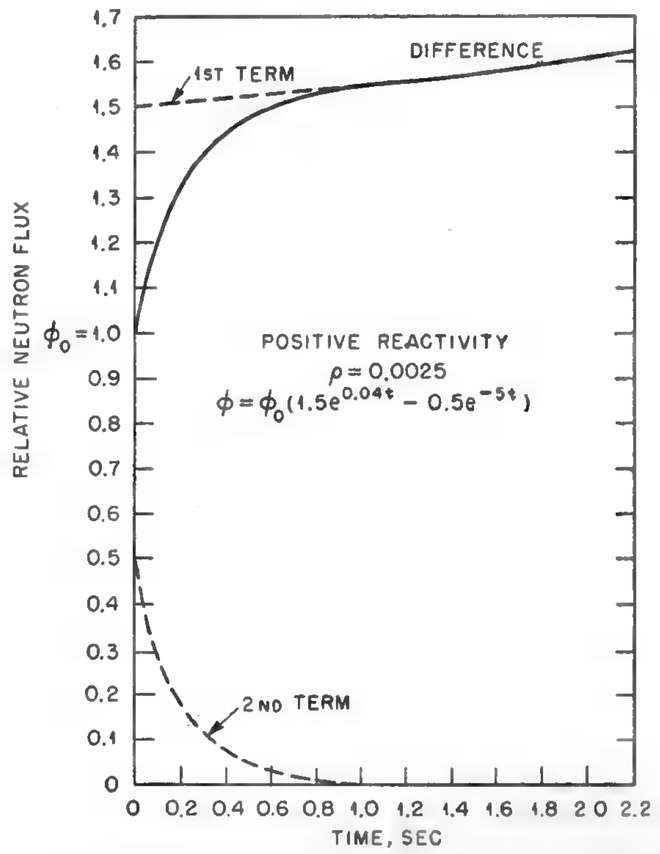


FIG. 4.1. Change of neutron flux with time for positive reactivity

* For a complete derivation of equation (4.25.2), see S. Glasstone and M. C. Edlund, "Elements of Nuclear Reactor Theory," D. Van Nostrand Co. Inc., 1952, ¶ 10.30 *et seq.*

Example The reactivity of a natural-uranium reactor, in which the thermal-neutron lifetime is 10^{-3} sec, is suddenly made 0.0025 positive; assuming one group of delayed neutrons, determine the subsequent change of the flux with time. Compare the stable period with that which would result if there had been no delayed neutrons.

For one (average) group of delayed neutrons λ is 0.08 sec^{-1}; ρ is given as 0.0025 and l as 10^{-3}, whereas β for uranium-235 is taken to be 0.0075. Consequently, by equation (4.25.2),

$$\phi = \phi_0(1.5e^{0.04t} - 0.5e^{-5.0t}).$$

The numerical values of each term on the right and of their differences are plotted in Fig. 4.1. The second (transient) term is seen to die out after about 0.8 sec.

By equation (4.20.1) the stable period $(1/\omega_0)$ is $1/0.04 = 25$ sec, assuming one group of delayed neutrons. If there had been no delayed neutrons the period, according to equation (4.9.2), would have been $10^{-3}/0.0025 = 0.4$ sec. The effect of the delayed neutrons in increasing the reactor period, i.e., in decreasing the rate of growth of the neutron flux after a change in the multiplication, is very marked.

4.27. A more complete calculation, taking into account all the groups of delayed neutrons, gives a stable period of 14 sec for a positive reactivity of 0.0025. The difference between this value and that obtained above (25 sec) arises largely from the approximations made in deriving equation (4.25.2). These approximations are much less serious for small reactivities, e.g., 0.001 or less, and then the results obtained using one group of delayed neutrons are reasonably good.

4.28. For very small reactivities, i.e., for very long periods, ρ in equation (4.26.2) may be neglected in comparison with β; this equation then becomes

$$T \approx \frac{\beta}{\lambda \rho}. \tag{4.28.1}$$

Since β/λ has a definite value for a given fissionable material, e.g., 0.092 for uranium-235 (see Table 4.1), it is seen that at low reactivities the stable reactor period is inversely proportional to the reactivity. As will be mentioned later, this fact has been utilized in connection with large reactors; for such reactors the effective multiplication factor is never much greater than unity and the reactivities are usually small.

4.29. It will be noted from Fig. 4.1 that immediately after the step change in the multiplication factor, i.e., at times very soon after $t = 0$, the neutron flux increases rapidly; this is sometimes referred to as the initial prompt rise of the flux. The rate of the initial rise may be calculated by first differentiating equation (4.24.1) with respect to time and setting $t = 0$, i.e.,

$$\left(\frac{\partial \phi}{\partial t}\right)_{t=0} = A_0\omega_0 + A_1\omega_1.$$

Upon introducing the values of A_0, A_1, ω_0, and ω_1 derived above, it is found that

$$\frac{1}{\phi_0} \cdot \frac{\partial \phi_0}{\partial l} = \frac{\lambda \rho \beta}{(\beta - \rho)^2} + \frac{\rho}{l}. \tag{4.29.1}$$

From the definition of the reactor period, e.g., equation (4.9.1),

$$\frac{1}{\phi} \cdot \frac{d\phi}{dt} = \frac{1}{T}$$

and if T_0 represents the initial reactor period at $t = 0$, it follows from equation (4.29.1) that

$$\frac{1}{T_0} = \frac{\lambda\rho\beta}{(\beta - \rho)^2} + \frac{\rho}{l}. \tag{4.29.2}$$

4.30. For moderately small values of ρ, the first term on the right side of equation (4.29.2) may be neglected, and then

$$T_0 \approx \frac{l}{\rho} \approx \frac{l}{k_{ex}}, \tag{4.30.1}$$

the final result being based on the fact that ρ is equal to k_{ex}/k_{eff}, by definition, whereas k_{eff} is not very different from unity. It is seen from equation (4.30.1) that the initial reactor period T_0 is identical with that given by equation (4.9.2) for the case where all the fission neutrons are released promptly. Thus, for small reactivities, the initial rate of growth of the neutron flux, following a step change, is almost the same as if all the fission neutrons were prompt.*

4.31. The physical interpretation of this situation is as follows. The number of delayed neutrons contributed to the chain reaction at any instant is proportional to the thermal-neutron flux at some previous time, but the number of delayed-neutron precursors produced by fission, i.e., the number of neutrons being delayed , is proportional to the flux at the given instant. For a short time after the change in the effective multiplication factor the neutron flux will not change very greatly, provided the reactivity is small. The number of delayed neutrons contributed to the system will then not be appreciably less than those being delayed. Hence, for a very short time, during the initial prompt rise, the reactor behaves as if all the fission neutrons were prompt, and the flux increases with a period of approximately l/k_{ex}. However, as the neutron flux grows with time, fewer delayed neutrons enter the chain-reacting system than are being delayed, and the rate of increase of the flux gradually falls off. Ultimately, the rate of growth is determined by the stable period, equal to $1/\omega_0$.

One Group of Delayed Neutrons: Negative Reactivity

4.32. The approximations made in deriving equation (4.25.2) are applicable for almost any negative values of ρ, especially for large negative values of the reactivity. This equation may thus be used to provide an indication of the manner in which the neutron flux decreases as the result of a negative step change in the effective multiplication factor. For purposes of comparison with the results obtained in the example given above, it will be supposed that ρ is -0.0025; taking the values of l, λ, and β to be the same as before, equation (4.25.2) now becomes

$$\phi = \phi_0(0.75e^{-0.02t} + 0.25e^{-10t}).$$

The values of the two terms and their sum are plotted in Fig. 4.2. It will be noted that the coefficients of both terms are now positive and the exponents are both negative. However, the large negative exponent in the second term on the right makes this damp out fairly rapidly, in about 0.5 sec, as seen in Fig. 4.2. When this term ceases to be significant, the stable period appears to be $-1/0.02$, i.e., -50 sec, compared to roughly 25 sec when ρ is 0.0025 positive (§ 4.27).

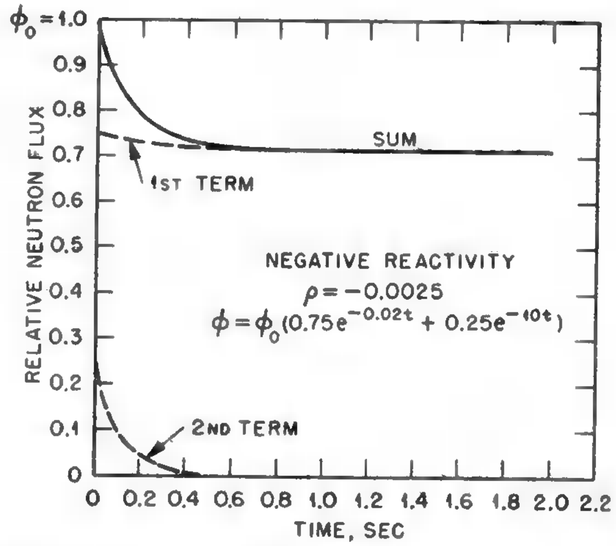


Fig. 4.2. Change of neutron flux with time for negative reactivity

4.33. If all the fission neutrons were prompt the reactor periods would be -0.4 sec for $\rho = -0.0025$ and 0.4 sec for $\rho = 0.0025$. It is evident, therefore, that, whereas the presence of delayed neutrons slows down the rate of increase of flux when the reactivity is positive, it slows down the rate of decrease to an even greater extent when the reactivity is negative. This may be seen more clearly from Fig. 4.3 which covers a considerably longer period of time than the two preceding figures.* It will be shown below that it is not possible to reduce the neutron flux in a reactor more rapidly than is permitted by the most-delayed neutron group (§ 4.41).

4.34. Since the arguments leading to equation (4.30.1) apply irrespective of the sign of the reactivity, it follows that immediately after the multiplication

* It should be understood that T_0 does not represent a stable reactor period. An examination of the derivation given above shows that it is a purely transient period arising from the second term on the right of equation (4.24.1).

* These curves are based on calculations using one (average) group of delayed neutrons. For the particular value of the positive reactivity chosen, i.e., 0.0025, the results, except at small times, are too low (see Isbin and Gorman, *loc. cit.*), although they are qualitatively correct.

factor of the reactor undergoes a step decrease, the initial period is equal to l/k_{ex}, where k_{ex} is now negative. The rate of decrease of the neutron flux is then the same as if all the neutrons were prompt. Thus, for very small times after the change in the multiplication factor, the two curves in Fig. 4.3 are seen to be symmetrical. The physical basis of this result is the same as given above for the case of positive reactivity. After a short time, however, the delayed neutron effect causes a considerable slowing down of the rate of decrease of flux.

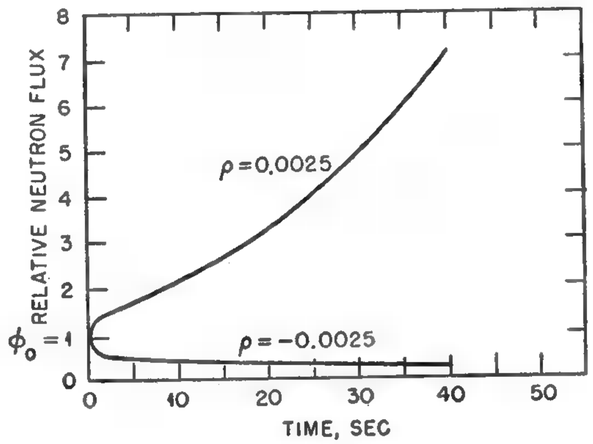


FIG. 4.3. Comparison of effects of positive and negative reactivities

REACTIVITY AND PERIOD: POSITIVE REACTIVITIES

4.35. If ω in equation (4.17.2) is replaced by $1/T$, where T is the stable reactor period, the result is

$$\rho = \frac{l}{Tk_{\text{eff}}} + \sum_{i=1}^{m} \frac{\beta_i}{1 + \lambda_i T}, \tag{4.35.1}$$

which provides a relationship between the reactivity and the stable period. If the period T is given, the reactivity can be calculated, since l, β_i and λ_i may be regarded as known, and k_{eff} may be taken as unity, for the results derived above apply only to systems that are not far from critical. The reverse procedure, i.e., the calculation of the stable reactor period for a given reactivity involves solution of an $(m + 1)$th, i.e., sixth, order equation. However, for positive reactivities that are not too high, results in good agreement with those given by this elaborate procedure can be obtained by the method based on one average group of delayed neutrons, as already described.

4.36. Considerable simplification is possible when the stable period is large, that is to say, when the reactivity is very small. In this case, unity may be neglected in comparison with each $\lambda_i T$ in the terms of the summation in equation (4.35.1). Further, the first term on the right will be small in comparison with the summation, so that for small reactivities, or large reactor periods, equation

(4.35.1) reduces to

$$\rho \approx \frac{1}{T} \sum_{i=1}^{m} \frac{\beta_i}{\lambda_i}$$

or

$$T \approx \frac{1}{\rho} \sum_{i=1}^{m} \frac{\beta_i}{\lambda_i}. \tag{4.36.1}$$

Since all the β_i's and λ_i's have definite values for a particular fissionable material, it is apparent that the reactivity and stable reactor period are inversely proportional to each other. The proportionality constant should be the same for all thermal reactors using a given fissionable material, and should be independent of the reactor size and structure, provided the reactivity is small.

4.37. It will be noted that equation (4.36.1) becomes identical with equation (4.28.1), based on a single group of delayed neutrons, provided the average decay constant λ is defined by

$$\lambda \equiv \beta / \sum_{i=1}^{m} \frac{\beta_i}{\lambda_i}.$$

This is, in fact, the definition used in § 4.22, so as to make the single group roughly equivalent to the m actual groups of delayed neutrons.

4.38. For small reactivities, the stable period for a given reactivity is seen to be independent of the neutron lifetime, l, because the first term on the right of equation (4.35.1) is then small in comparison with the second. However, for larger values of ρ, i.e., for shorter reactor periods, the effect of the neutron life-

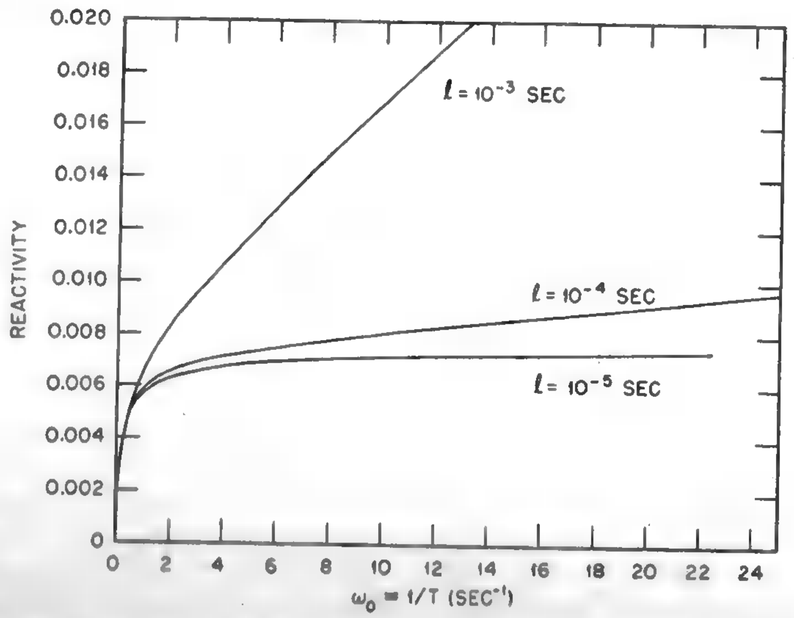


FIG. 4.4. Reciprocal of stable period for various neutron lifetimes

time becomes evident. The relationship between the reactivity and $1/T$, the reciprocal of the stable period, is shown in Fig. 4.4 for neutron lifetimes of 10^{-3}, 10^{-4}, and 10^{-5} sec. The stable periods for a few reactivity values are also given in Table 4.2, for the same three neutron lifetimes. At low reactivities, the

TABLE 4.2. REACTOR PERIOD AND REACTIVITY

Neutron Lifetime (sec)	10^{-3}	10^{-4}	10^{-5}
Reactivity	Reactor Period (sec)		
0.001	60	60	60
0.003	10	10	10
0.005	2.5	2.5	2.5
0.007	0.8	0.2	0.14
0.010	0.3	0.04	0.0003

three curves coincide, so that for a specified reactivity the period is independent of the neutron lifetime, as stated above. For higher reactivities, e.g., greater than about 0.005, the stable period corresponding to a given reactivity decreases as the lifetime decreases. This means that for higher reactivities the neutron flux will increase at a more rapid rate the smaller the neutron lifetime. At such reactivities, reactors with short neutron lifetimes, e.g., fast reactors, are more difficult to control than are those in which the neutrons have longer lifetimes. At low reactivities, however, no difference should be experienced with respect to control.

4.39. At sufficiently high values of ρ, so that T is small, the second term on the right side of equation (4.35.1) can be neglected in comparison with the first. The stable period for large reactivities is then given by

$$T = \frac{l}{\rho k_{\text{eff}}} = \frac{l}{k_{\text{ex}}},$$

which is the same as if all the fission neutrons were prompt. Thus, when the reactivity is small, the rate of increase of neutron flux (after the transients have died out), for a given value of ρ, is determined essentially by the neutron delay time, and is independent of the much shorter neutron lifetime. At high reactivities, however, particularly when $\rho > \beta$, the contribution of the delayed neutrons is negligible in comparison with that of the prompt neutrons. The reactor then behaves as if virtually all of the fission neutrons were prompt; the stable period is then dependent on the neutron lifetime, as shown earlier.

Reactivity and Period: Negative Reactivities

4.40. For numerically small negative values of the reactivity, the approximations made in deriving equation (4.28.1) are valid. The negative stable period

is then inversely proportional to the negative reactivity. However, for large negative reactivities, the stable periods calculated on the basis of one average group of delayed neutrons, which are fairly satisfactory for positive reactivities, are too small. This is because, after the transients have died out, the rate of change of neutron flux, especially for moderate and large negative reactivities, is determined by the most-delayed neutrons, and not by the average, as is essentially true for positive reactivities.

4.41. In physical terms, it may be said that when the reactivity is made negative, as in reactor shut-down, delayed neutrons, resulting from fissions occurring before shut-down, continue to be released. These prevent the neutron flux from decreasing as rapidly as would have been the case if there were no delayed neutrons. After a little time, the shorter-lived precursors will have decayed almost completely, and the delayed neutrons remaining are essentially those belonging to the group having the precursor of longest life, i.e., the one for which λ is 0.0125 sec^{-1} (see Table 4.1). In these circumstances, equation (4.35.1) simplifies to

$$\rho \approx \frac{\beta}{1 + \lambda_1 T},$$

where λ_1 has the value just given. It should be noted that β is larger than usual since the delayed neutrons now constitute a greater proportion of the fission neutrons. As ρ increases numerically, it is evident that $(1 + \lambda_1 T) \to 0$, so that, for large negative reactivities, the stable period, T, approaches $-1/\lambda_1$, i.e., $-1/0.0125 = -80$ sec.

4.42. Observations have shown that thermal reactors containing carbon (graphite) or ordinary water as moderator do, in fact, attain a stable period of about -80 sec when shut down (see Fig. 5.13). When the moderator is beryllium or heavy water (deuterium oxide), the period is even longer due to the fact that gamma rays from the fission products continue to release neutrons as a result of the photoneutron reaction with the moderator (§ 2.74). Allowance for the effect of the photoneutrons can be made by introducing appropriate values for β and λ into the kinetic equations.*

The Inhour Formula

4.43. The reactivity is sometimes expressed in terms of the inverse hour or "inhour" unit, defined as the reactivity which will make the stable reactor period equal to 1 hour, i.e., 3600 sec. Thus, the value of the inhour unit in terms of reactivity is obtained by setting T in equation (4.35.1) equal to 3600 sec, since the λ_i are usually expressed in sec^{-1}. The reactivity of a reactor in inhours, represented by Ih, is then obtained upon dividing equation (4.35.1) by the corresponding value of the inhour unit; thus,

* Cf. A. Lundby, *Nucleonics*, 11, No. 4, 16 (1953).

$$\text{Ih} = \frac{\dfrac{l}{Tk_{\text{eff}}} + \displaystyle\sum_{i=1}^{m} \frac{\beta_i}{1 + \lambda_i T}}{\dfrac{l}{3600k_{\text{eff}}} + \displaystyle\sum_{i=1}^{m} \frac{\beta_i}{1 + 3600\lambda_i}}. \tag{4.43.1}$$

This expression is the correct form of the inhour formula, although the related equations (4.17.2) and (4.35.1) are sometimes referred to by this name.

4.44. It can be seen from equation (4.43.1) that, in general, the reactivity given in inhours does not define the stable reactor period in a simple manner. Nevertheless, for large reactors, such as the natural uranium-graphite reactors, the reactivity is frequently expressed in inhours. The reason is that for small reactivities, i.e., for long reactor periods, unity may be neglected in comparison with $\lambda_i T$ and $3600\lambda_i$, and equation (4.43.1) then reduces to

$$\text{Ih} \approx \frac{3600}{T} \quad \text{or} \quad T \approx \frac{3600}{\text{Ih}}, \tag{4.44.1}$$

so that the reactivity in inhours is inversely related to the stable reactor period in a simple manner. In fact, if the reactor period is expressed in hours, it is equal to the reciprocal of the reactivity in inhours.

4.45. By comparing equations (4.36.1) and (4.44.1), a relationship between ρ and Ih may be obtained, namely,

$$\frac{1}{\rho} \sum_{i=1}^{m} \frac{\beta_i}{\lambda_i} = \frac{3600}{\text{Ih}}.$$

For uranium-235, the sum of the five β_i/λ_i values is 0.092, and so

$$\rho = 2.6 \times 10^{-5}\text{Ih}.$$

Although this result is strictly applicable only for long reactor periods, nevertheless it is sometimes combined with equation (4.35.1) to give the relationship

$$\text{Ih} \approx \frac{1}{2.6 \times 10^{-5}} \left(\frac{l}{T} + \sum_{i=1}^{m} \frac{\beta_i}{1 + \lambda_i T} \right),$$

where k_{eff} has been taken as unity; l and T are usually expressed in seconds, and so the λ_i's are in reciprocal seconds. Upon inserting the known values of β_i and λ_i, from Table 4.1, and that of l for thermal neutrons in the given reactor, an expression is obtained relating the reactivity in inhours to the reactor period.

The Prompt-critical Condition

4.46. When a reactor is critical on prompt neutrons alone, it is said to be *prompt critical*. The required condition for prompt criticality can be obtained from equation (4.14.1). If the delayed-neutron source term is ignored, so that prompt fission neutrons only are considered, this equation becomes

$$[k_{\text{eff}}(1 - \beta) - 1]\phi = l \frac{d\phi}{dt}.$$

For the reactor to be critical, the flux must not vary with time, i.e., $d\phi/dt$ must be zero; since ϕ and l are not zero, the prompt-critical condition is

$$k_{\text{eff}}(1 - \beta) - 1 = 0$$

or

$$\frac{k_{\text{eff}} - 1}{k_{\text{eff}}} = \beta. \tag{4.46.1}$$

By the definition of equation (4.17.1), the left side is equal to the reactivity, so that the required condition is

$$\rho = \beta,$$

i.e., a reactor becomes prompt critical when the reactivity is equal to the fraction of delayed neutrons. A thermal reactor in which the fissionable material is uranium-235 thus becomes prompt critical when ρ is 0.0075, so that k_{eff}, which is equal to $1/(1 - \beta)$ by equation (4.46.1), is then close to 1.0075.*

4.47. When the effective multiplication factor is greater than the prompt-critical value, the chain reaction can be maintained on the prompt neutrons alone. The reactor is then said to "outrun" the delayed neutrons. If the delayed neutrons made no contribution at all, it can be readily shown that the stable reactor period would be given by

$$T = \frac{l}{k_{\text{ex}}(\text{prompt})}, \tag{4.47.1}$$

where $k_{\text{ex}}(\text{prompt})$ is the amount by which the effective multiplication factor exceeds the prompt-critical value, i.e.,

$$k_{\text{ex}}(\text{prompt}) = k_{\text{eff}} - \frac{1}{1 - \beta} \approx k_{\text{eff}}(1 - \beta) - 1,$$

since $1 - \beta$ is not greatly different from unity. Further, by definition, k_{eff} is equal to $1/(1 - \rho)$, as seen from equation (4.17.1), and so it follows that

$$k_{\text{ex}}(\text{prompt}) \approx \frac{\rho - \beta}{1 - \rho} = (\rho - \beta)k_{\text{eff}}.$$

The stable period for reactivities *in excess* of the prompt-critical value is then given by equation (4.47.1) as

$$T \approx \frac{l(1 - \rho)}{\rho - \beta} = \frac{l}{k_{\text{eff}}} \cdot \frac{1}{\rho - \beta}. \tag{4.47.2}\dagger$$

* Although the data in Tables 2.11 and 4.1 indicate that β is 0.0073, it is taken here to be 0.0075 for the reason given in § 2.167.

† The same expression can be obtained directly from equation (4.35.1) by neglecting $\lambda_i T$ in comparison with unity, since the reactor period is short. The treatment given above, however, brings out the physical significance of the result.

This result is applicable only when $\rho > \beta$; it does not hold at prompt critical for then $\rho - \beta$ is zero, and the stable period can be obtained only by solving equation (4.35.1).

4.48. If $\rho - \beta$ is large enough for β to be neglected in comparison with ρ, equation (4.47.2) reduces to

$$T \approx \frac{l}{\rho k_{\text{eff}}} = \frac{l}{k_{\text{ex}}},$$

as given in § 4.39. The reactor period is then approximately equal to that which would result if all the neutrons were prompt. The contribution of the delayed fission neutrons is negligible in comparison with that of the prompt neutrons.

4.49. Some indication of the rate of increase in the neutron flux as a result of fission of uranium-235, for various reactivities, will be found in Fig. 4.5; the

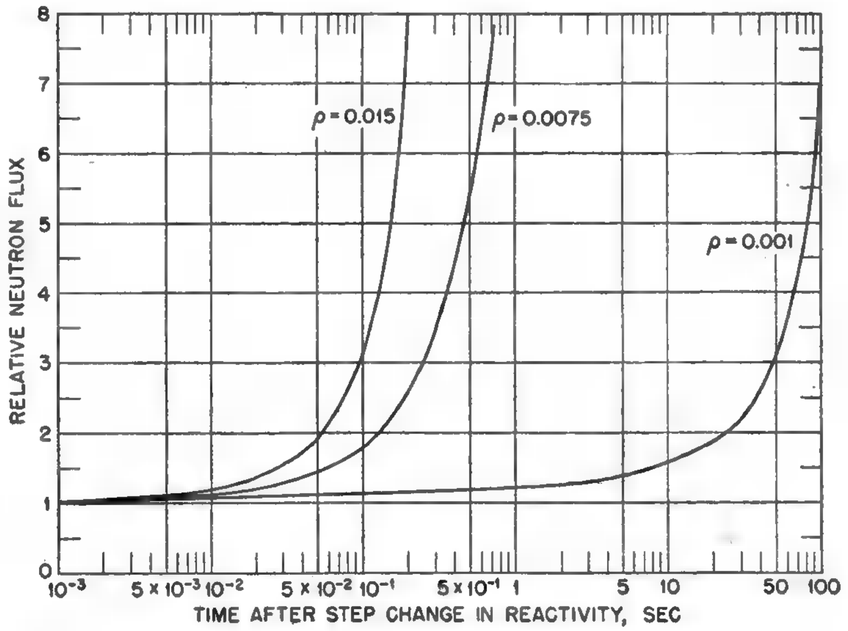


FIG. 4.5. Rate of increase of neutron flux following a sudden increase in reactivity

values of ρ are 0.001; 0.0075, i.e., prompt critical; and 0.015, respectively, the neutron lifetime l being taken as 10^{-3} sec in each case. It should be noted that in order to plot the three curves on one figure, the time scale has been made logarithmic; the change in behavior as ρ is increased is thus more marked than would appear at first sight. An examination of the curves shows that when ρ is 0.001 it takes over 100 sec for the neutron flux to increase 10-fold, but when ρ is 0.0075, the same increase occurs in about 1 sec, and when ρ is 0.015, it would require about 0.2 sec. The rapid increase in the flux when the reactivity exceeds the prompt-critical value would make the reactor difficult to control, and special precautions must be taken that this condition does not arise in reactor operation.

4.50. On account of its special significance, the prompt-critical condition is sometimes used as the basis for a reactivity unit, especially for reactors having considerable latent (or built-in) excess multiplication (see Chapter VI). The unit, called "the dollar," is defined by

$$\text{Reactivity in dollars} \equiv \frac{k_{\text{ex}}}{\beta}.$$

A "cent" represents a one-hundredth part of a dollar. When prompt critical, a reactor has a reactivity of one dollar. For a thermal reactor using uranium-235 as the fissionable material, for example, a reactivity of \$1.50 would imply an excess multiplication of 1.50×0.0075, i.e., 0.0113, and an effective multiplication factor of 1.0113.*

Linear Change of Multiplication Factor

4.51. In the discussion so far it has been postulated that the multiplication factor of the reactor undergoes a sudden (or step) change. A more difficult problem arises when the change is a linear function of the time, sometimes referred to as a "ramp" change, when the effective multiplication factor is expressed by

$$k_{\text{eff}} = k_0 + rt, \tag{4.51.1}$$

where r is a constant. This type of change is an approximation to that which occurs when a control rod of a reactor is withdrawn or inserted at a constant rate, over a small range. Some simplification in the representation is possible if the neutron density, rather than the neutron flux, is used in the kinetic equations, and so the appropriate changes will first be made.

4.52. If ϕ in equation (4.14.1) is replaced by nv, where n is the neutron density and v the velocity of the thermal neutrons, and recalling the definitions of k_{eff} and l as given by equations (3.125.1) and (3.127.1), it can be readily shown that

$$[k_{\text{eff}}(1 - \beta) - 1]\frac{n}{l} + pe^{-B^2\tau}\sum_{i=1}^{m}\lambda_i C_i = \frac{\partial n}{\partial t}. \tag{4.52.1}$$

In an analogous manner, equation (4.10.1) can be transformed into

$$\frac{\partial C_i}{\partial t} = \beta_i \frac{k_{\text{eff}}}{pe^{-B^2\tau}} \cdot \frac{n}{l} - \lambda_i C_i. \tag{4.52.2}$$

Upon substituting the expression for $\lambda_i C_i$ obtained from equation (4.52.2) into (4.52.1) the result is

$$(k_{\text{eff}} - 1)\frac{n}{l} - pe^{-B^2\tau}\sum_{i=1}^{m}\frac{\partial C_i}{\partial t} = \frac{\partial n}{\partial t}. \tag{4.52.3}$$

4.53. The equation (4.51.1), representing the time variation of the multiplication factor, can now be inserted for k_{eff} in equation (4.52.3); if this is then

* J. P. Frass, Report AECD-3260.

differentiated with respect to t, and $\partial^2 C_i/\partial t^2$ eliminated by means of equations (4.52.1) and (4.52.2), it is found that

$$\frac{d^2n}{dt^2} + \left[\lambda + \frac{1 - k_0(1 - \beta)}{l} - \frac{r(1 - \beta)}{l}\, t\right]\frac{dn}{dt} + \left[\frac{\lambda(1 - k_0) - r(1 - \beta)}{l} - \frac{r\lambda t}{l}\right] n = 0, \qquad (4.53.1)$$

assuming the delayed neutrons to consist of a single group with an average decay constant λ. This is of the form of

$$\frac{d^2n}{dt^2} + (A + Bt)\frac{dn}{dt} + (C + Dt)n = 0,$$

where the values of A, B, C, D are determined by the initial condition of the reactor and the rate of the linear change of multiplication.

4.54. Several solutions of the differential equation (4.53.1) have been given, but they are too complex to be reproduced here.* Nevertheless, linear changes in the effective multiplication factor, following upon different operating conditions, are important in reactor operation; a few of the possibilities are summarized in Table 4.3. The columns headed k_{eff}, n, and dn/dt refer to the conditions before the linear change is made in k_{eff}, i.e., for $t < 0$; the last column gives the sign of r, i.e., of dk_{eff}/dt, for the linear change in k_{eff} when $t > 0$. The term "period safety" implies the automatic insertion of a control rod when the reactor period exceeds a prescribed value (see Chapter VI).

TABLE 4.3. CONDITIONS FOR LINEAR CHANGE IN REACTIVITY

	$k_{\text{eff}}(t < 0)$	$n(t < 0)$	$\frac{dn}{dt}(k < 0)$	$r(k > 0)$
Start-up	<1	small	zero	positive
Shut-down	~1	operating level	zero	negative
Recovery	<1	(not important)	negative	positive
Period safety	>1	(not important)	positive and large	negative

Kinetics of Circulating-fuel Reactors

4.55. In some reactors (or reactor designs) a fluid fuel, either a solution, a slurry, or a melt, is circulated through a heat exchanger outside the reactor to remove the heat generated by fission. In such a system some of the delayed fission neutrons are released outside the reactor core and so do not contribute to the fission chain. As far as the neutron flux (or density) variation with time is concerned, equations of the form of (4.14.1) or (4.52.1) are still applicable, but equations (4.10.1) or (4.52.2) for the concentrations of the delayed-neutron

* See R. R. Coveyou and T. W. Mulliken, Report AECD-2407; S. Wallach, Report WAPD-18; H. Hurwitz, Report AECD-2438; *Nucleonics*, **5**, No. 1, 61 (1949).

precursors must be modified to allow for the fact that some of these precursors decay outside the reactor. Further, another expression must be introduced to describe the rate of decrease in the concentration of the precursors in the heat exchanger. The resulting differential equations are more complicated than those given above, but they can be solved in certain cases.* Without going into details, it can be seen that the result of circulating the fuel will be to decrease the effective value of β, the fraction of delayed neutrons released in the reactor, but to increase the effective decay constant, λ, since the most delayed neutrons will tend to be released outside the reactor. The over-all result, as may be seen from equation (4.26.2), for example, will be to decrease the reactor period corresponding to a given change in reactivity; this is, of course, to be expected. In general, the effects due to delayed neutrons will be less marked in a circulating-fuel reactor than in one in which the fuel remains within the reactor core.

Solution of Kinetic Equations by Reactor Simulators

4.56. Solution of the differential equations encountered in reactor kinetics has been greatly facilitated by the use of electronic simulators. A simulator, in general, is a "real time" analogue computer; that is to say, it is a device which displays the analytical behavior of the system *on the same time scale* as would apply to the system itself. Thus a simulator can take the place of the simulated system, such as a reactor, in relation to other "real" systems. For example, if it is required to test a servomechanism to be employed in conjunction with a particular reactor, a simulator may be used for the purpose. Frequently, it is found convenient to alter the time scale of the computer; it then ceases to be a simulator in the strict sense of the term and is merely an analogue computer. Although such a computer is useful for analytical purposes, it cannot be used for testing servomechanisms and other systems.

4.57. With the correct conversion factors, a simulator will act in a manner that is essentially identical with the actual system. There are, however, certain limitations upon the accuracy of the analysis performed with the aid of a simulator. In the first place, although the simulator can be designed to obey a set of differential equations, it is doubtful whether the behavior of the simulated system is represented exactly by these equations. As will be seen below, it is possible to design an electrical simulator which satisfies the equations of reactor kinetics, but it is known that these equations are based on numerous approximations. Consequently the reactor simulator cannot provide a complete representation of the actual behavior of the reactor. Further, it is not possible to take into account transient or spatial variations of a relatively minor character which must inevitably occur within an active system, such as a reactor, of appreciable size. In spite of their obvious limitations, simulators have proved very useful in the study of reactor kinetics. The results they give are at least as good as those that could be obtained in a more laborious manner by direct analysis.

* Cf. J. A. Fleck, Jr., *Nucleonics*, **12**, No. 10, 52 (1954).

4.58. Three basic electrical elements are used in the construction of computers; these are the resistor, the capacitor, and the inductor. The currents (I) through these elements are different functions of the voltage (E) across them. Thus, in the case of a resitance R, the current is given by $I = E/R$; for a capacitance C, the result is $I = C(dE/dt)$; and for an inductance L, the value of I is $\frac{1}{L}\int E\,dt$. Consequently the three elements can be used for different operations. Pure inductances, free from resistance and capacitance, are difficult to obtain and so they are avoided as far as possible. In some cases, mechanical devices are employed in conjunction with electrical elements (§ 4.112).

4.59. The basic principles of a simulator for reactor kinetics can be explained most simply by considering, in the first place, one group of delayed neutrons. In this case, equation (4.23.1) is applicable, and if this is multiplied through by ϕ, the thermal-neutron flux, the result, after slight rearrangement, is

$$\frac{\beta\omega}{\omega + \lambda}\phi + \frac{l\omega}{k_{\text{eff}}}\phi - \rho\phi = 0. \tag{4.59.1}$$

In view of the significance of ω as given by equation (4.15.1) or (4.19.1), it is evident that ω, as an operator, is equivalent to d/dt; hence, bearing this in mind, equation (4.59.1) is seen to be the differential equation, in operational form, representing the time variation of neutron flux in a reactor of reactivity ρ.*

4.60. Consider, next, the electrical circuit shown in Fig. 4.6. The direct-coupled amplifier is "double-ended," so that the positive voltage ($+E$) output is equal in magnitude but opposite in sign to the negative voltage ($-E$). Using

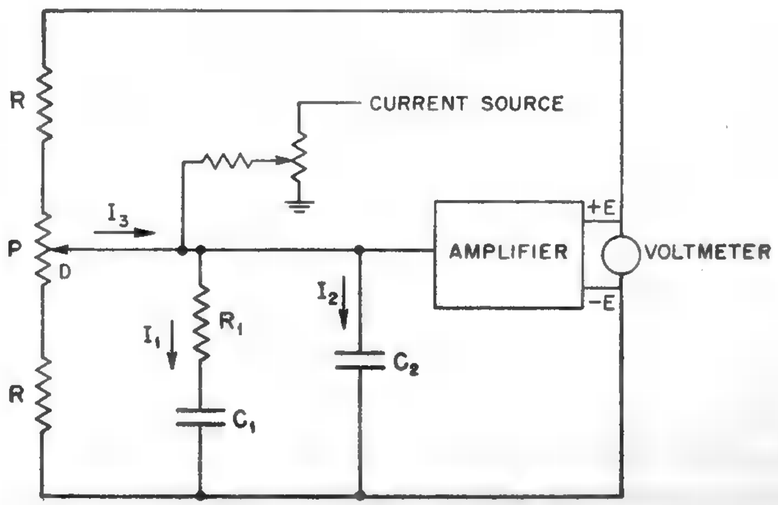


FIG. 4.6. Simplified circuit of reactor simulator

* It is of interest to note that equation (4.59.1) or, more correctly, the form involving m groups of delayed neutrons, which is equivalent to equation (4.17.2), can be readily derived from equations (4.10.1) and (4.14.1) by using ω to represent the operation d/dt.

ω in place of d/dt, the currents I_1, I_2, and I_3 through the various parts of the circuit are given by

$$I_1 = \frac{C_1\omega}{R_1C_1\omega + 1}E,$$

$$I_2 = C_2\omega E,$$

and

$$I_3 = f(D, P, R)E,$$

where $f(D, P, R)$ is a simple function of the fixed resistances R and P, and of the variable distance D at which the arm of the potentiometer P makes contact. According to Kirchhoff's second law

$$I_1 + I_2 - I_3 = 0,$$

and so it follows that the differential equation satisfied by the circuit under consideration is

$$\frac{C_1\omega}{R_1C_1\omega + 1}E + C_2\omega E - f(D, P, R)E = 0. \tag{4.60.1}$$

4.61. An examination of equations (4.59.1) and (4.60.1) shows that they are analogous, and so the circuit of Fig. 4.6 can evidently form the basis of a simulator for reactor kinetics. An exact equivalence can evidently be obtained by the choice of R_1 and C_1 in terms of λ and β, of C_2 in terms of l/k_{eff}, and P and R so that the distance D is a measure of the reactivity. In order that the capacitance C_2 may have a definite value, it is necessary to make an approximation in equation (4.59.1) by supposing that k_{eff} is close to unity. Thus, by choosing the proper components of the circuit, the output voltage E of the amplifier becomes a direct measure of the neutron density (or flux). By changing the position of the contact of the potentiometer P, so as to correspond to a particular reactivity, e.g., for a step or ramp change in the multiplication factor, the time variation of the neutron density can be observed from the readings of a suitable voltmeter placed across the output of the amplifier.

4.62. In a reactor simulator of the form just described, it is a simple matter to take into account the m, i.e., five, groups of delayed neutrons. All that is necessary is to use m resistance-capacitance delay networks, such as R_1-C_1 in Fig. 4.6. The differential equation obeyed by the electrical system will then contain m terms similar to the first on the left of equation (4.60.1), one corresponding to each group of delayed neutrons. Allowance for all these groups, instead of one average group, makes possible a more exact representation of transient behavior. This is important in connection with the testing of servomechanisms for automatic reactor control.*

* For descriptions of reactor kinetics simulators, see P. R. Bell and H. A. Strauss, *Rev. Sci. Inst.*, **21**, 760 (1950); W. Pagels, *Proc. Am. Inst. Eng.*, **70**, 1422 (1951); F. E. O'Meara, *J. Appl. Phys.*, **24**, 1200 (1953); F. E. O'Meara and K. H. Fischbeck, *Proc. Conf. on Nuc. Eng.*, Berkeley, 1953, D-20.

Sinusoidal Variation of Reactivity

4.63.* In the dynamic analysis of any system, e.g., in connection with the design of a servomechanism, it is necessary to determine the response or transfer function.† This involves a knowledge of the change resulting from a small variation, often sinusoidal in character, in one of the parameters of the system referred to by the general name of *parametric forcing*. In the case of a reactor, for example, this might be represented by the change in the neutron density due to a small harmonic variation in the reactivity, i.e., reactivity forcing.

4.64. Suppose that a reactor is operating in a steady state, so that $n = n_0$, $C_i = C_{i0}$, and $k_{\text{eff}} = k_0$, and let k_{eff} undergo small variations, such that $k_{\text{eff}} = k_0 + k'$, where k_0 is constant and k' is small and time dependent. Similarly, let the values of n and C_i corresponding to the new values of k_{eff} be $n = n_0 + n'$ and $C_i = C_{i0} + C_i'$. Upon substituting these expressions for k_{eff}, n, and C_i into equations (4.52.1) and (4.52.2), and neglecting all products of primed quantities, since they will be very small, the results are

$$\frac{dn'}{dt} + \frac{1 - k_0(1 - \beta)}{l} n' = \frac{n_0(1 - \beta)}{l} k' + pe^{-B^2\tau} \sum_{i=1}^{m} \lambda_i C_i' \qquad (4.64.1)$$

and

$$\frac{dC_i'}{dt} = \frac{\beta_i(n_0 k' + n' k_0)}{lpe^{-B^2\tau}} - \lambda_i C_i'. \qquad (4.64.2)$$

These equations will hold for any small variation in k_{eff}.

4.65. In the particular case where the variations are sinusoidal in character with a forcing frequency of ω_f, e.g., for sinusoidal reactivity forcing, let

$$k' = k_a e^{j\omega_f t}$$

and, correspondingly,

$$n' = n_a e^{j\omega_f t} \quad \text{and} \quad C_i' = C_{ia} e^{j\omega_f t},$$

where k_a, n_a, and C_{ia} are the respective complex amplitudes. The operator d/dt can thus be replaced by $j\omega_f$, and hence it can be found from equations (4.64.1) and (4.64.2) that

$$\frac{n_a}{k_a} = \frac{n'}{k'} = n_0 \frac{1 - \beta + \sum_{i=1}^{m} \frac{\beta_i \lambda_i}{j\omega_f + \lambda_i}}{1 - (1 - \beta)k_0 + j\omega_f l - k_0 \sum_{i=1}^{m} \frac{\beta_i \lambda_i}{j\omega_f + \lambda_i}}.$$

In the case of one (average) group of delayed neutrons, this would reduce, after

* The material for §§ 4.63 to 4.74 was contributed by W. H. Jordan and J. D. Trimmer.

† Cf. G. S. Brown and D. P. Campbell, "Principles of Servomechanisms," John Wiley & Sons, Inc., 1948; G. J. Thaler and R. G. Brown, "Servomechanism Analysis," McGraw-Hill Book Co. Inc., 1953; or other standard textbook. See also, J. M. Harrer, *et al.*, *Nucleonics*, 10, No. 8, 32 (1952).

rearrangement, to

$$n_a \frac{1 - k_0}{n_0 k_a} = \frac{1 + j\omega_f \left(\frac{1 - \beta}{\lambda}\right)}{1 - \omega_f^2 \frac{l}{\lambda(1 - k_0)} + \frac{j\omega_f}{\lambda}\left(1 + \frac{\beta k_0 + l\lambda}{1 - k_0}\right)}. \qquad (4.65.1)$$

4.66. An equation of this form is satisfied by the electrical circuit in Fig. 4.7; if E_i is the input voltage and E_o the output, then the voltage ratio is given by

$$\frac{E_o}{E_i} = \frac{1 + j\omega_f C R_0}{1 - \omega_f^2 LC + j\omega_f C(R + R_0)}, \qquad (4.66.1)$$

using $j\omega_f$, as above, to represent the operator d/dt. It is evident, from a comparison of equations (4.65.1) and (4.66.1), that the circuit in Fig. 4.7 can form the basis of a simulator to represent the behavior of a nuclear reactor undergoing a small sinusoidal change in reactivity. If the input voltage, E_i, is made

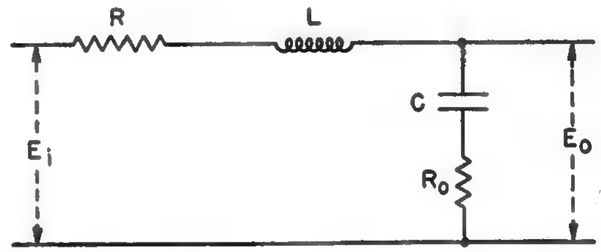


Fig. 4.7. Electrical circuit for determining effects of sinusoidal reactivity variations

equivalent to $n_0 k_a/(1 - k_0)$, where k_a is the complex amplitude of the effective multiplication factor, then the output voltage E_o will be equivalent to n_a, the corresponding neutron density amplitude. Thus, equation (4.66.1) can indicate the response of a reactor to reactivity forcing, and the circuit can be used in an electrical simulator for testing a system to be used with the reactor near an equilibrium condition, e.g., in testing a servomechanism to be used for automatic control at a prescribed power level.

Kinetics of Reactor with Primary Source

4.67. In the discussion so far, the possibility of a primary (or extraneous) neutron source, i.e., a neutron source not part of the fission chain, being present in the reactor has been disregarded. All reactors actually have such a source, even if it is due only to spontaneous fission and to the action of cosmic rays. Some reactors have, in addition, a built-in neutron source, of the type described in Chapter II, to facilitate start-up operations (see § 6.161). When the reactor is operating at a reasonable power level, the contribution of the primary source is negligible, as shown in § 3.197, and the effective multiplication factor in the stationary state is essentially unity. At very low neutron fluxes, however, such

as might arise in reactor start-up and in certain experiments, the contribution of the source may be significant.

4.68. Although the diffusion equation (4.2.1) is quite general, it was assumed in its subsequent development that no primary source was present in the reactor. If there is a constant source, represented by S_0 neutrons/(cm³)(sec), equation (4.52.3), for example, would become

$$(k_{\text{eff}} - 1)\frac{n}{l} - pe^{-B^2\tau}\sum_{i=1}^{m}\frac{\partial C_i}{\partial t} + S_0 = \frac{\partial n}{\partial t}. \tag{4.68.1}$$

If the reactor is in a steady state, so that $\partial C_i/\partial t$ and $\partial n/\partial t$ are both zero, it follows from equation (4.68.1) that

$$n = \frac{lS_0}{1 - k_{\text{eff}}}, \tag{4.68.2}$$

so that if n is to be finite, k_{eff} must be less than unity. A reactor containing a primary source will thus be stable in a subcritical state, i.e., with $k_{\text{eff}} < 1$. It is seen, too, that the neutron density (or flux) is determined, apart from l, by the source S_0 and the value of $1 - k_{\text{eff}}$. With a truly critical reactor, i.e., $S_0 = 0$ and $k_{\text{eff}} = 1$, however, the neutron density in the steady state can have any value, in principle.

4.69. It may be noted, in passing, that equation (4.68.2) can be used to derive the expression for the subcritical multiplication given in § 3.192. In a non-multiplying medium, i.e., in the absence of fissionable material, k_{eff} is zero and so the neutron density, according to equation (4.68.2), will be proportional to S_0. On the other hand, in a subcritical multiplying medium, the neutron density is determined by $S_0/(1 - k_{\text{eff}})$. The multiplication, i.e., the ratio of the neutron density (or flux) due to a given source in a multiplying medium to the density (or flux) due to the same source in the absence of fissionable material, is thus $1/(1 - k_{\text{eff}})$, in agreement with equation (3.192.1).

4.70. A further difference between a reactor with a primary source and a critical reactor is their behavior when k_{eff} is changed. For a critical reactor, an increase in k_{eff} will mean that the neutron flux will increase indefinitely, but if a source S_0 is present, it is apparent from equation (4.68.2) that, provided k_{eff} is less than unity, the neutron density will merely increase until a new equilibrium value is attained. Of course, if k_{eff} were made greater than unity, the flux would increase without limit. For example, if k_{eff} is 0.90, the value of n will be $10lS_0$; then, if k_{eff} undergoes a step change to 0.99, the neutron density will increase until it reaches $100lS_0$, where it remains constant. Similarly, if S_0 were changed, the reactor would still be stable at a new equilibrium state. A comparison of the behavior of reactors with a primary source ($S_0 > 0$) and without such a source ($S_0 = 0$) as a result of changes either in the effective multiplication factor or the source is given in Table 4.4. The entry at the bottom of the third column

TABLE 4.4. COMPARISON OF REACTORS WITH AND WITHOUT PRIMARY SOURCE

	Reactor with Source	*Reactor without Source*
Stability condition	$k_{\text{eff}} < 1$	$k_{\text{eff}} = 1$
Change in multiplication	Stable at new flux	Unstable
Change in source	Stable at new flux if $k_{\text{eff}} < 1$. Unstable if $k_{\text{eff}} \geqslant 1$.	Unstable

implies that the critical reactor would become unstable if a source were introduced.

4.71. It is of interest to determine the rate of increase of the neutron flux accompanying an increase in the effective multiplication factor of a subcritical reactor with a primary source. The results are important in the start-up of a reactor, before it becomes critical. From equation (4.68.1) it follows that

$$n = \frac{l}{k_{\text{eff}} - 1}\left(pe^{-B^2\tau}\sum_{i=1}^{m}\frac{\partial C_i}{\partial t} - \frac{\partial n}{\partial t}\right) + \frac{lS_0}{1 - k_{\text{eff}}}. \tag{4.71.1}$$

If there were no source present, the solution of this equation for n would be the sum of $m + 1$ exponentials, analogous to equation (4.18.1), and so in the present case, it is evident that

$$n = \frac{lS_0}{1 - k_{\text{eff}}} + A_0e^{\omega_0 t} + A_1e^{\omega_1 t} + \cdots + A_me^{\omega_m t}, \tag{4.71.2}$$

where A_0, A_1, etc., are determined by $m + 1$ initial conditions; these are obtained by setting $t = 0$ in equation (4.71.2) and $\partial C_i/\partial t = 0$ in the m equations (4.10.1) to give n_0 and the m values of C_{i0}, respectively. The relationship between ω_1,

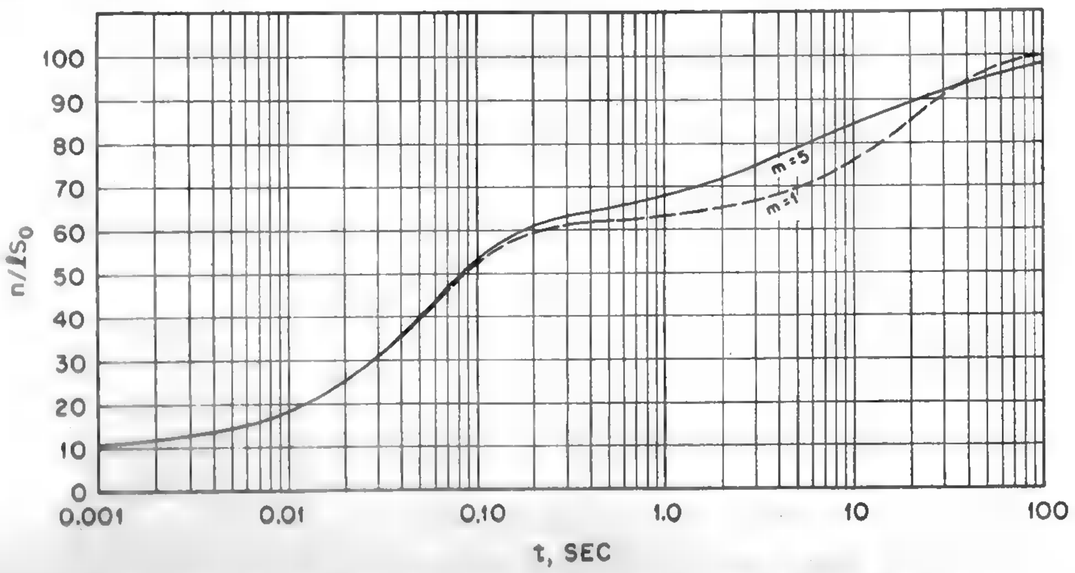


FIG. 4.8. Effect of sudden increase in multiplication of subcritical reactor

etc., and k_{eff} (or ρ) is not derived so simply as when no source is present but when k_{eff} is not much less than unity, the values of ω to be used in equation (4.71.2), for a given ρ (or k_{eff}), may be obtained from equation (4.17.2).

4.72. The results of a calculation made in the manner just described are shown in Fig. 4.8. It is based on the supposition that the reactor, with source S_0, has been in a steady state with $k_{\text{eff}} = 0.90$, when, at $t = 0$, the value of k_{eff} is suddenly increased to 0.99.* Thus, the initial value of n (at $t = 0$) is given by equation (4.68.2) as $10lS_0$, and the final steady-state neutron density is $100lS_0$. In evaluating the ω's, $\rho = (k_{\text{eff}} - 1)/k_{\text{eff}}$ is -0.0101, and l is taken as 10^{-3} sec. The first term on the right of equation (4.71.2) is $100lS_0$ and all the other terms have negative coefficients and negative exponents. Hence, as t increases, the exponential terms decrease, so that n approaches the limiting value $100lS_0$. The full curve in Fig. 4.8 is based on five groups of delayed neutrons, and the broken curve on one average group ($\lambda = 0.08$ sec^{-1}); the agreement is, on the whole, very good.

Sinusoidal Variation of Primary Source

4.73. The problem of variations in the primary source, i.e., source forcing, in a subcritical reactor is not of great immediate importance, but the subject will be considered here partly for the sake of completeness and partly because it is not impossible that it may have ultimate practical application. Suppose there is a sinusoidal variation in the source strength, so that

$$S = S_0 e^{j\omega_f t}$$

and the corresponding values of n and C_i are

$$n = n_a e^{j\omega_f t} \quad \text{and} \quad C_i = C_{i0} e^{j\omega_f t}.$$

The effective multiplication factor remains constant at k_{eff}. If these expressions for n and S (in place of S_0) are inserted into equation (4.71.1), and the result is combined with equation (4.10.1), for one average group of delayed neutrons, with $j\omega_f$ replacing the operator d/dt, it is found that

$$n_a \frac{1 - k_{\text{eff}}}{lS_0} = \frac{1 + j\omega_f/\lambda}{1 - \omega_f^2 \dfrac{l}{\lambda(1 - k_{\text{eff}})} + \dfrac{j\omega_f}{\lambda}\left(1 + \dfrac{l\lambda + \beta k_{\text{eff}}}{1 - \beta k_{\text{eff}}}\right)}.$$

If this result is compared with equation (4.66.1), it will be apparent that the circuit in Fig. 4.7 can be used as a simulator for a subcritical reactor subject to source forcing. The input voltage, E_i, is equivalent to $lS_0/(1 - k_{\text{eff}})$, and the output voltage, E_o, represents the corresponding amplitude of the neutron density, n_a.

4.74. A summary of the electrical analogues of various reactor parameters for both reactivity forcing and source forcing is given in Table 4.5. Amplitude and

* The calculations involve the implicit assumption that $k_{\text{eff}} = 0.90$ is sufficiently close to unity for the results derived above to apply; this is probably true for $k_{\text{eff}} = 0.90$, at least.

TABLE 4.5. ELECTRICAL ANALOGUES OF REACTOR QUANTITIES

Electrical Parameters	Reactor Parameters	
	Reactivity Forcing	Source Forcing
Input voltage, E_i	$\dfrac{n_0 k_a}{1 - k_0}$	$\dfrac{lS_0}{1 - k_{\text{eff}}}$
Output voltage, E_o	n_a	n_a
Time constant CR_0	$(1 - \beta)/\lambda$	$1/\lambda$
Time constant $\sqrt{LC}$	$\sqrt{l/[\lambda(1 - k_0)]}$	$\sqrt{l/[\lambda(1 - k_{\text{eff}})]}$
Damping ratio $\dfrac{R + R_0}{2\sqrt{L/C}}$	$\dfrac{1}{2}\left[\sqrt{\dfrac{1 - k_0}{l\lambda}} + \sqrt{\dfrac{l\lambda}{1 - k_0}} + \dfrac{\beta k_0}{\sqrt{l\lambda(1 - k_0)}}\right]$	$\dfrac{1}{2}\left[\sqrt{\dfrac{1 - k_{\text{eff}}}{l\lambda}} + \sqrt{\dfrac{l\lambda}{1 - k_{\text{eff}}}} + \dfrac{\beta k_{\text{eff}}}{\sqrt{l\lambda(1 - k_{\text{eff}})}}\right]$
Resistance ratio R/R_0	$\dfrac{l\lambda + \beta}{(1 - \beta)(1 - k_0)}$	$\dfrac{l\lambda + \beta k_{\text{eff}}}{1 - k_{\text{eff}}}$

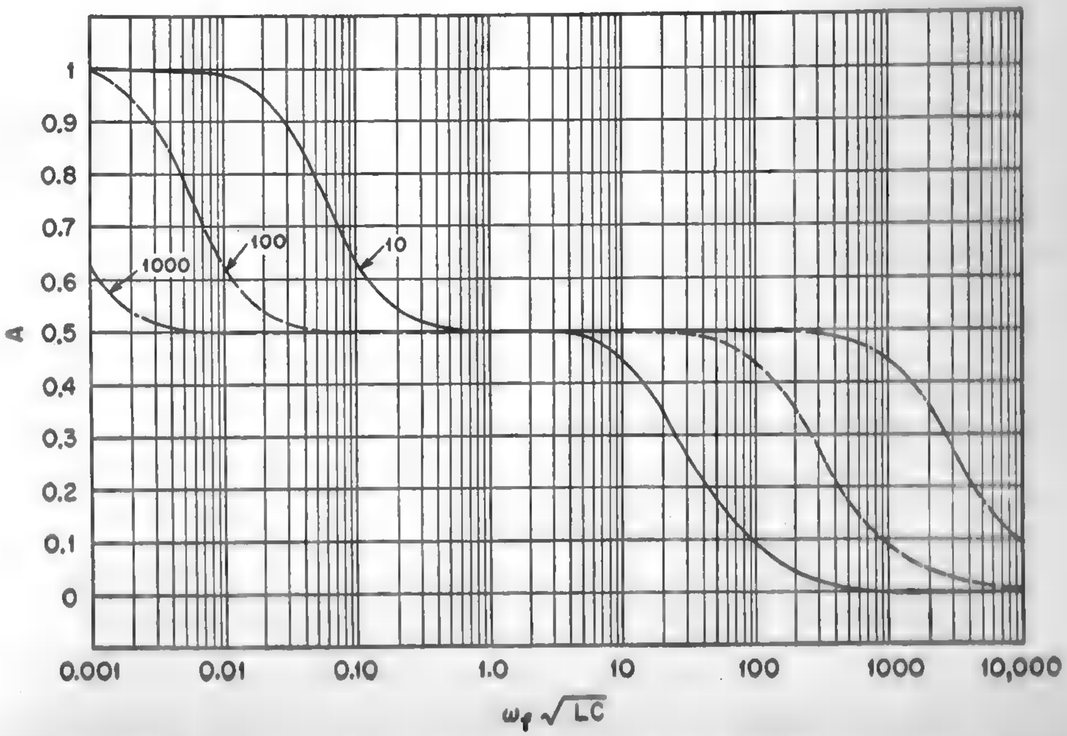


FIG. 4.9. Amplitude ratio as function of frequency for sinusoidal forcing

phase characteristics, based on the approximation of one group of delayed neutrons, are plotted in Figs. 4.9 and 4.10 in terms of electrical parameters, so that they are applicable, in general, to both types of forcing. The amplitude ratio A and the phase angle φ are defined by setting $E_o/E_i = Ae^{j\varphi}$. These quantities are important in determining the transfer function to be used in the design and testing of equipment, such as servomechanisms, associated with reactor operation and control. The curves are given for three different values of the damping

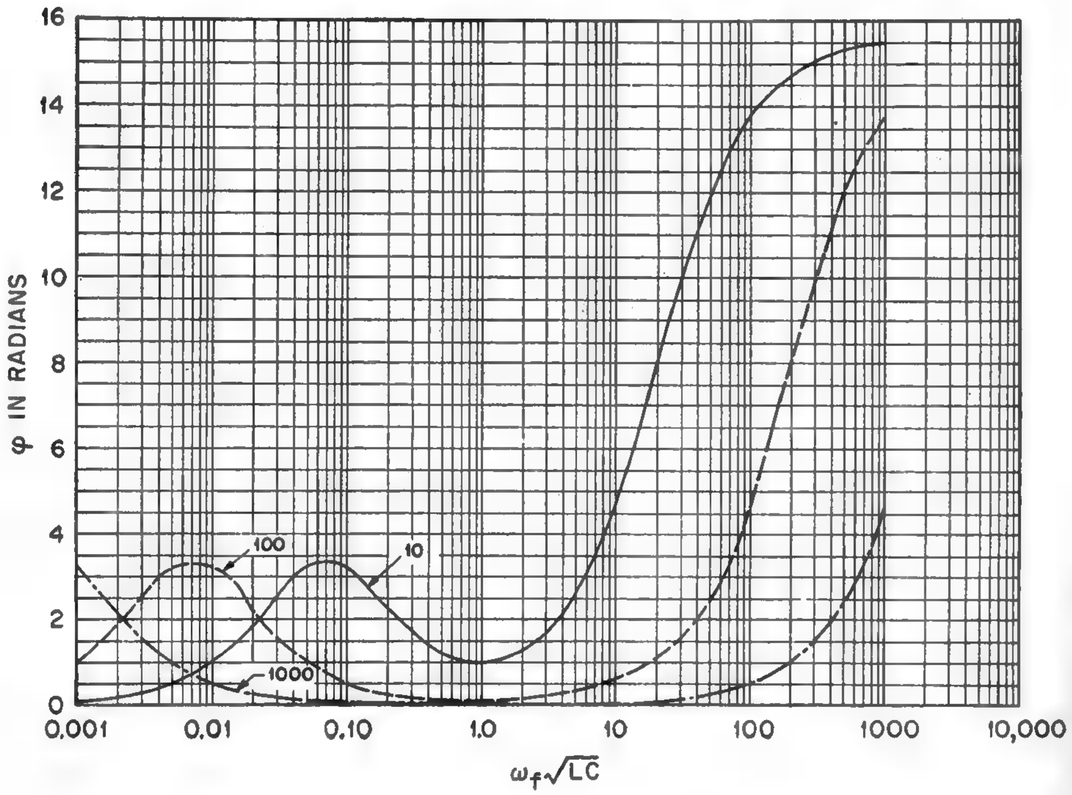


FIG. 4.10. Phase angle as function of frequency for sinusoidal forcing

ratio, namely, 10, 100, and 1000; because of the nature of the reactor parameters, the electrical quantities would always be such as to make the damping overcritical, i.e., the damping ratio is greater than unity. For simplicity, R has been taken equal to R_0 in the calculations.

EFFECT OF TEMPERATURE ON REACTIVITY

General Considerations

4.75. An important cause of transient changes in the effective multiplication factor in an operating reactor is variation in the temperature of the system. Such variations may be localized, e.g., due to nonuniformity of structure affecting the flow of coolant at particular points, or they may affect the reactor as a whole. A change in the rate of flow, for example, would gradually alter the temperature of the reactor. A similar effect would result from a change in the power demand, i.e., in the heat removed from the coolant, so that the temperature at which the latter enters the reactor would be altered. Whether such temperature changes cause the multiplication factor of the reactor to increase or decrease is of great practical significance.

4.76. A change of temperature will alter the multiplication factor, or reactivity, for at least two reasons. First, the mean energy of the thermal neutrons is changed and, as a result, the various scattering and absorption cross sections will be affected; and second, there will be changes in the diffusion length, in the Fermi age, and in the reactor size due to alteration in the density of the reactor materials. For the theoretical treatment, to be outlined below, it is convenient to separate the effect of temperature on reactivity into two main parts: the *nuclear temperature coefficient*, which is determined by the effect on the nuclear cross sections, and the *density temperature coefficient*, due to changes in density (and volume) of the system. The discussion which follows applies particularly to a homogeneous reactor system, but the general conclusions will hold, to a good approximation, for most heterogeneous reactors.

Nuclear Temperature Coefficients

4.77. In view of the definition of k_{eff}, as given by equation (3.123.2), it follows that for a large reactor,

$$k_{\text{eff}} \approx \frac{k}{1 + B^2(L^2 + \tau)} = \frac{k}{1 + M^2B^2},$$

where $M^2 = L^2 + \tau$ is the migration area of the neutrons (§ 3.99). Upon rearrangement, and introducing the definition of the reactivity, ρ, it is found that

$$\rho = \frac{k - 1}{k} - \frac{B^2(L^2 + \tau)}{k} = \frac{k - 1}{k} - \frac{B^2M^2}{k}. \tag{4.77.1}$$

At constant density the buckling, B^2, which is a geometrical quantity, will not be affected, and so the nuclear temperature coefficient of reactivity will be determined by changes in L^2, τ, and k.

4.78. In the thermal-energy region, absorption cross sections may be assumed to follow the $1/v$ law (§ 2.171). Since the kinetic energy ($\frac{1}{2}mv^2$) of a neutron is proportional to the absolute temperature, T, this means that thermal cross sections will vary as $1/T^{\frac{1}{2}}$. If σ_a is the absorption cross section at temperature T and σ_{a0} is the value at T_0, then

$$\sigma_a = \sigma_{a0}\left(\frac{T_0}{T}\right)^{\frac{1}{2}}. \tag{4.78.1}$$

Scattering cross sections do not change greatly with temperature, and for the present purpose it is sufficient to suppose that they remain constant.

4.79. According to equation (3.38.1), the square of the diffusion length is related to the macroscopic absorption and scattering cross sections by

$$L^2 = \frac{1}{3\Sigma_a\Sigma_s}, \tag{4.79.1}$$

provided the medium does not absorb too strongly and scattering is isotropic in the laboratory system. Ignoring, for the present, the effect of density changes on the number of nuclei per cm³, which is involved in the macroscopic cross sections, it is evident from equations (4.78.1) and (4.79.1) that

$$L^2 = L_0^2\left(\frac{T}{T_0}\right)^{\frac{1}{2}}, \tag{4.79.2}$$

since Σ_s may be regarded as being independent of temperature.

4.80. In view of the definition of the Fermi age, as given by equation (3.90.1), it is apparent that the temperature variation of τ will be determined by the effect on D/Σ_s, i.e., on $1/\Sigma_s^2$, since D is equal to $1/3\Sigma_s$, assuming isotropic scattering. Since it has been postulated that, apart from the effect of density, Σ_s is independent of temperature, the same applies to the Fermi age.

4.81. The next point to consider is the result of changes in cross sections on k, the infinite multiplication factor. If all the absorbers present, including fuel, moderator, coolant, poisons, etc., obey the $1/v$ law, then the thermal utilization, f (§ 3.133), will not be affected by temperature. The resonance escape probability may decrease somewhat as the temperature is raised, because there is an increase in the effective resonance integral (§ 3.135), largely due to the Doppler effect referred to in § 4.83. On the whole, however, since η and ϵ (§ 3.131) remain unchanged, it may be assumed here, as a reasonable approximation, that k is independent of temperature. But this will not be the case if the average neutron energy is in the vicinity of a resonance of one or more of the reactor components, for then the $1/v$ law will not be obeyed.

4.82. Returning now to equation (4.77.1), inserting equation (4.79.2) for L^2 and noting that τ and k are unaffected by temperature, as far as nuclear cross sections are concerned, the result is

$$\rho = \frac{k-1}{k} - \frac{B^2}{k}\left[L_0^2\left(\frac{T}{T_0}\right)^{\frac{1}{2}} + \tau\right].$$

The nuclear temperature coefficient, at constant density, i.e., the temperature coefficient of reactivity due only to changes in cross section, assuming general applicability of the $1/v$ law, is then obtained upon differentiation with respect to T; thus,

$$\left(\frac{\partial\rho}{\partial T}\right)_d = -\frac{B^2L_0^2}{2k}\frac{1}{(TT_0)^{\frac{1}{2}}},$$

where the subscript d indicates constant density.* The temperature coefficient

* Because ρ is used to represent reactivity, the symbol d is employed for density in this chapter.

at the temperature T is obtained by setting $T_0 = T$, so that

$$\left(\frac{\partial\rho}{\partial T}\right)_d = -\frac{B^2L^2}{2kT}. \tag{4.82.1}$$

Since B^2, L^2, k, and T are all positive quantities, it is apparent that the nuclear temperature coefficient must be negative. Therefore, as far as changes in cross section are concerned, an increase of temperature will result in a decrease in the reactivity and, hence, in the effective multiplication factor. The change in the resonance escape probability, which has been neglected, will probably operate in the same direction.

4.83. The foregoing treatment is based on the assumption that the absorption cross sections of all substances present in the reactor obey the $1/v$ law. This is, of course, not always true. A special case would arise if there happened to be a sharp resonance just above the normal thermal range; an increase of temperature might then bring the average neutron energy into the resonance region. This result would be favored by the so-called neutron Doppler effect, which is a broadening of the resonance with increasing temperature due to the increased random motion of the target nuclei. The consequent increased neutron absorption would make a marked negative contribution to the nuclear temperature coefficient. Any nonfissionable material present in the reactor could cause this to happen by decreasing the thermal utilization.

4.84. It should be borne in mind that the effect of operation near a resonance does not necessarily always lead to a negative temperature coefficient of reactivity. If an increase of temperature brings the average neutron energy closer to the resonance of the fissionable material, the result would be an increase in the thermal utilization and in the infinite multiplication factor. A similar effect can be due to a decrease in parasitic capture of neutrons as a consequence of moving away from the resonance of a poison or other nonfissionable substance as the temperature is increased.

Density Temperature Coefficients

4.85. Change of temperature affects the density of the reactor materials, and this alters the reactivity in two ways. First, by changing the number of nuclei per cm³, it alters the macroscopic cross sections, apart from any changes in the microscopic cross sections considered above. Second, since there is an over-all change in the size of the reactor, the buckling is altered.

4.86. The number of nuclei per unit volume is directly proportional to the density, and so all macroscopic cross sections vary in a similar manner. It can be shown, therefore, from the definitions of L^2 and τ, that these quantities are inversely proportional to the square of the density. In a homogeneous system, where density changes have the same effect on all the components, it is possible to write

$$L^2 + \tau = L_0^2\left(\frac{d_0}{d}\right)^2 + \tau_0\left(\frac{d_0}{d}\right)^2,$$

or

$$M^2 = M_0^2 \left(\frac{d_0}{d}\right)^2,$$

where M^2 and d are the migration area and density at the absolute temperature T, and M_0^2, and d_0 are the values at T_0. If this is substituted into equation (4.77.1), the result is

$$\rho = \frac{k-1}{k} - \frac{B^2 M_0^2}{k}\left(\frac{d_0}{d}\right)^2.$$

If the buckling is taken to be constant, as well as the microscopic cross sections, as stated above, then

$$\left(\frac{\partial \rho}{\partial T}\right)_{B^2,\,\sigma} = \frac{2B^2 M_0^2}{k} \cdot \frac{d_0^2}{d^3} \cdot \frac{\partial d}{\partial T}. \tag{4.86.1}$$

4.87. If α is the coefficient of *linear* expansion of the reactor materials, then $V = V_0[1 + \alpha(T - T_0)]^3$ and

$$d = \frac{d_0}{[1 + \alpha(T - T_0)]^3},$$

so that

$$\frac{\partial d}{\partial T} = -\frac{3\alpha d_0}{[1 + \alpha(T - T_0)]^4}.$$

Insertion of this result into equation (4.86.1) then gives

$$\left(\frac{\partial \rho}{\partial T}\right)_{B^2,\,\sigma} = -\frac{6B^2 M_0^2}{k}\left(\frac{d_0}{d}\right)^3 \frac{\alpha}{[1 + \alpha(T - T_0)]^4}.$$

At $T_0 = T$, this becomes

$$\left(\frac{\partial \rho}{\partial T}\right)_{B^2,\,\sigma} = -\frac{6B^2 M^2 \alpha}{k},$$

or since $1 + M^2B^2 \approx k$ for a (large) critical reactor,

$$\left(\frac{\partial \rho}{\partial T}\right)_{B^2,\,\sigma} = -\frac{6\alpha(k-1)}{k}. \tag{4.87.1}$$

Because $k - 1$ and α are positive this contribution to the density temperature coefficient is negative.

4.88. The reactivity change due to an over-all change in reactor size arises from the effect on the buckling. Differentiation of equation (4.77.1) with respect to B, assuming the other parameters to be constant, gives

$$\left(\frac{\partial \rho}{\partial B}\right)_{\sigma} = -\frac{2BM^2}{k} = -\frac{2(k-1)}{kB}, \tag{4.88.1}$$

since $M^2 = (k-1)/B^2$. In the case of a *spherical reactor*, $B = \pi/R$ (Table 3.7) and $R = R_0[1 + \alpha(T - T_0)]$, where R is the radius; hence,

$$\frac{dR}{dT} = \alpha R_0$$

and, at T,

$$\frac{dB}{dT} = -\frac{\pi}{R^2} \cdot \frac{dR}{dT} = -\frac{\alpha\pi}{R}. \tag{4.88.2}$$

Consequently, from equations (4.88.1) and (4.88.2), and the fact that $B = \pi/R$,

$$\left(\frac{\partial \rho}{\partial T}\right)_{\sigma} = \frac{\partial \rho}{\partial B} \cdot \frac{\partial B}{\partial T} = \frac{2\alpha(k-1)}{k}. \tag{4.88.3}$$

This result, derived for a spherical reactor, may be regarded as being generally applicable. Since $k - 1$ and α are positive, this part of the density temperature coefficient differs from that considered above in being positive.

4.89. The density effect on the multiplication factor is generally small. If the reactor is of such a type that the ratio of fissionable material to moderator is not affected by density changes, e.g., a uranium-graphite reactor, then k will also be unaffected. If the fuel elements are solid and the moderator is a liquid, such as heavy water, maintained at constant level, a change in density of the liquid will alter the fuel-to-moderator ratio. However, since the thermal utilization and the resonance escape probability will usually change in opposite directions (cf. § 3.177, *et seq.*), the over-all effect on k is likely to be small.

4.90. A change in density of coolant can have an important influence on the infinite multiplication factor. As a rule, a decrease in density due to an increase of temperature will mean that the mass of coolant present in the reactor will decrease. If the coolant is a fairly good neutron absorber, as is the case with ordinary water, for example, the decrease in mass, i.e., in the number of capturing nuclei, will result in an increase in the multiplication factor. The contribution of the coolant to the density temperature coefficient of reactivity will then be positive.

Example A natural uranium-graphite reactor, operating at a mean temperature of about 120°C (400°K), has an effective multiplication factor of 1.06. Estimate the temperature coefficient of reactivity of this reactor.

For graphite L_m^2 at the absolute (Kelvin) temperature 400°K is roughly 2850 cm², and by equation (3.147.1), which also applies to a heterogeneous reactor, L^2 for the reactor is 2850 $(1 - f)$, where f, the thermal utilization, is about 0.9 (see Table 3.9); hence, the value of L^2 to be used here is 285 cm². The age, τ, of thermal neutrons in the reactor approaches 400 cm², so that $M^2 = L^2 + \tau$ may be taken as 680 cm². This result, in combination with $k = 1.06$, gives 8.8×10^{-5} cm^{-2} for B^2, since $k \approx 1 + M^2B^2$. The coefficient of linear expansion, α, may be taken as 10^{-5} per °C, this being a reasonable average for graphite and uranium. Using these values and $T = 400$°K, it is found, from equations (4.82.1), (4.87.1), and (4.88.3), that

$$\left(\frac{\partial \rho}{\partial T}\right)_{d} = -\frac{B^2 L^2}{2kT} = -2.8 \times 10^{-5} \text{ per °C (or } -1.55 \times 10^{-5} \text{ per °F)}$$

$$\left(\frac{\partial \rho}{\partial T}\right)_{m,\,\sigma} = -\frac{6\alpha(k-1)}{k} = -0.34 \times 10^{-5} \text{ per °C (or } -0.19 \times 10^{-5} \text{ per °F)}$$

$$\left(\frac{\partial \rho}{\partial T}\right)_{\sigma} = \frac{2\alpha(k-1)}{k} = 0.11 \times 10^{-5} \text{ per °C (or } 0.06 \times 10^{-5} \text{ per °F)},$$

so that, as far as these three terms are concerned, the net temperature coefficient of reactivity is -3.0×10^{-5} per °C, i.e., -1.7×10^{-5} per °F. (This is almost exactly the temperature coefficient of the X-10 reactor at Oak Ridge, but the close agreement is partly fortuitous.)

4.91. Reactors moderated by ordinary or heavy water have larger negative temperature coefficients than do graphite reactors; this is mainly due to the temperature coefficient of expansion of water being nearly ten times as great as that of graphite. Another contributing factor is the generally higher value of $(k-1)/k$, which is about 0.06 in a natural uranium-graphite reactor but might be 0.2 for one having heavy water as moderator. Most reactors moderated by ordinary water, e.g., Water Boiler (or HRE) and Swimming Pool types, use highly enriched fuel (about 90 per cent uranium-235); such reactors have large negative temperature coefficients, e.g., -30×10^{-5} per °C (-17×10^{-5} per °F) for the Water Boiler and -14×10^{-5} per °C (-7.5×10^{-5} per °F) for the Swimming Pool type. These are accounted for partly by the high value of $(k-1)/k$, which is about 0.4, and partly by the temperature coefficient of expansion of water. Reactors of the Water Boiler type are generally designed so as to permit overflow of the fuel solution when it expands due to an increase in temperature. This has the effect of decreasing the amount of fissionable material in the reactor core and as a result there is a considerable negative contribution to the temperature coefficient of reactivity.

4.92. It should be understood that the foregoing remarks apply to reactors in which water is the moderator, although they will also be true for reactors in which water is both moderator and coolant. However, if the water acts almost exclusively as coolant (within the reactor core) while the neutron moderation takes place in another medium, e.g., heavy water (as in the Canadian NRX reactor), beryllium, or graphite, the magnitude of the negative temperature coefficient is decreased and it may even become positive. This is because the cooling water makes a positive contribution to the reactivity temperature coefficient for the reason given in § 4.90.

Temperature and Reactor Stability

4.93. The effect of temperature on the inherent stability of a reactor has two aspects, namely, static stability and dynamic stability. A reactor possesses *static stability* to temperature changes if it has a negative temperature coefficient of reactivity. If, for any cause, there is a rise of temperature within the reactor, the effective multiplication factor will then tend to decrease. As a result the rate of fission (and of heat release) will also decrease and this will offset the rise in temperature. If the reactivity temperature coefficient were positive, an increase of temperature in the reactor core would result in an increased fission rate

and a further rise of temperature. Such a reactor would be unstable to temperature and operation would be unsafe.

4.94. Since a considerable part of the reactivity temperature coefficient is due to density changes in the reactor, the response to an increase or decrease of temperature is not instantaneous. As a result of these delays oscillations may develop in the neutron flux and reactor power. A reactor is said to have good *dynamic stability* when these oscillations are rapidly damped out because of the inherent characteristics of the system. In most reactors the delayed fission neutrons make an important contribution to the dynamic stability. As seen earlier, the effect of these neutrons is to slow down both the rate of increase and of decrease of the neutron flux resulting from changes in the reactivity. A special situation arises in connection with circulating-fuel reactors because, as mentioned in § 4.55, some of the delayed fission neutrons are released outside the reactor core. However, theoretical considerations have shown that, with proper design, the loss in dynamic stability resulting from the decrease in the proportion of delayed neutrons can be largely compensated by the damping effect caused by the circulation of the fuel.* It is certainly a fact that the HRE, in which an aqueous fuel solution is circulated continuously through an external heat exchanger, has remarkable dynamic stability to temperature changes (§ 13.102).

FISSION PRODUCT POISONING

Effect of Poisons on Reactivity

4.95. During the course of operation of a nuclear reactor, the fission fragments and their many decay products accumulate. Among these substances there are some, xenon-135 and samarium-149, in particular, which have large cross sections for thermal-neutron absorption. These nuclei, therefore, act as reactor poisons and affect the multiplication factor, chiefly by decreasing the thermal utilization. The concentration of fission product poison in a reactor is related to the thermal-neutron flux. Consequently, when the reactivity is changed, so that there is an accompanying change in the flux, the concentration of fission product poison will be affected and this will, in turn, influence the reactivity. It would appear, therefore, that some allowance for the effect of poisoning should be included in the reactor kinetics equations considered in the early sections of this chapter. However, the rate of change of fission product concentration with time is, in general, small compared to that of the flux. Hence, the equations giving the time rate of change of neutron flux may be treated quite independently of those for the fission product poisons.

4.96. Although fission products thus have little direct effect on reactor kinetics, they may have an important influence on the reactivity, and this must be taken into consideration in reactor design. As seen in Chapter II, because

* W. K. Ergen, *J. Appl. Phys.*, **25**, 702 (1954).

of the opposing effects of formation, as a direct or indirect result of fission, and removal, by radioactive decay and by neutron capture, the concentration of a particular species will attain an equilibrium value while the reactor operates at a constant power level. When the reactor is shut down, however, the concentration of the fission product may increase to a maximum, as explained in § 2.198, *et seq.* Both the equilibrium amount during operation and the maximum after shut-down depend on the neutron flux (or power output) of the reactor. The effect on the reactivity of a particular fission product poison will thus depend on the conditions of operation of the reactor as well as on the nature of the poison.

4.97. Of the four factors which make up the infinite multiplication factor (§ 3.134), the thermal utilization is essentially the only one affected by the poison. Let f be the thermal utilization in a reactor without poison and let f' be the value with poison; assuming, for simplicity, that the system is homogeneous, then

$$f = \frac{\Sigma_u}{\Sigma_u + \Sigma_m} \quad \text{and} \quad f' = \frac{\Sigma_u}{\Sigma_u + \Sigma_m + \Sigma_p},$$

where Σ_u, Σ_m, and Σ_p are the macroscopic cross sections for the absorption of thermal neutrons in fuel, moderator, and poison, respectively. The *poisoning*, P, of a reactor has been defined as the ratio of the number of thermal neutrons absorbed by the poison to those absorbed in the fuel; hence, in the present case,

$$P \equiv \frac{\Sigma_p}{\Sigma_u}. \tag{4.97.1}$$

If the ratio Σ_u/Σ_m is represented by z, then the expressions for f and f' can be transformed to

$$f = \frac{z}{z+1}, \quad \text{and} \quad f' = \frac{z}{z+1+Pz}.$$

4.98. Because the concentration of fission product poison is generally very small, its effect on the nonleakage probability while slowing down is negligible. However, the presence of a strong absorber in the reactor will decrease the thermal-neutron diffusion length, since $L^2 = D/\Sigma_a$; consequently there will be some increase in the thermal nonleakage probability $1/(1 + L^2B^2)$. As $1 + L^2B^2$ is close to unity, at least in large reactors, the nonleakage probability is fairly insensitive to changes in L^2. For the present purpose, it is thus reasonable to make the approximation of taking the effective multiplication factor to be proportional to the thermal utilization, with or without poison.

4.99. Let the effective multiplication factor without poison be k_{eff} and that with poison k_{eff}'; then

$$\frac{k_{\text{eff}}' - k_{\text{eff}}}{k_{\text{eff}}'} = \frac{f' - f}{f'} = -\frac{Pz}{1+z}. \tag{4.99.1}$$

If the reactor without poison is just critical, so that k_{eff} is unity, the left side of

equation (4.99.1) becomes equivalent to the (negative) reactivity change due to the poison; thus

$$\rho = -\frac{Pz}{1+z}.$$

In an enriched reactor, in particular, z is large in comparison with unity, and so the reactivity decrease resulting from the presence of a poison is roughly equal to the poisoning, P, i.e., to the ratio of Σ_p to Σ_u.

Xenon Poisoning During Operation

4.100. The most important fission product poison is xenon-135 because of its exceptionally large absorption cross section — about 3.5×10^6 barns — for thermal neutrons. This isotope is formed to a small extent (0.3 per cent) as a direct product of fission, but the main proportion in a reactor originates from tellurium-135, produced in 5.6 per cent of the slow-neutron fissions of uranium-235. Tellurium-135 has a half life of only 2 min and undergoes a series of stages of negative beta decay, as follows:

$$\text{Te}^{135} \xrightarrow{2\text{m}} \text{I}^{135} \xrightarrow{6.7\text{h}} \text{Xe}^{135} \xrightarrow{9.2\text{h}} \text{Cs}^{135} \xrightarrow{2\times10^4\text{y}} \text{Ba}^{135} \text{ (stable)}.$$

Consequently iodine-135 is rapidly formed in appreciable amounts, and although it is not itself a strong neutron absorber, it decays to yield the poison, xenon-135.

4.101. On account of the short half life of tellurium-135, the analytical treatment of xenon poisoning may be simplified, without introducing appreciable error, by assuming that iodine-135 is produced directly in fission with a fractional yield of 0.056. According to the argument in § 2.193, the net rate of formation of iodine-135 may thus be represented by

$$\frac{dI}{dT} = -\lambda_I I - \sigma_I \phi I + \gamma_I \Sigma_f \phi,$$

where I is the concentration of the iodine, λ_I is its radioactive decay constant, σ_I is its thermal-neutron absorption cross section, and γ_I is the fission yield, i.e., 0.056; Σ_f is the macroscopic thermal fission cross section of the fuel material in the reactor, and ϕ is the thermal-neutron flux. This expression is, of course, identical with equation (2.187.1), and so the equilibrium concentration I_0 of iodine-135, attained after the reactor has been operating for some time at constant flux, is given by equation (2.188.1) as

$$I_0 = \frac{\gamma_I \Sigma_f \phi}{\lambda_I^*} \approx \frac{\gamma_I \Sigma_f \phi}{\lambda_I}. \tag{4.101.1}$$

Since σ_I is quite small, λ_I^*, defined by equation (2.188.2) as $\sigma_I\phi + \lambda_I$, may be replaced by λ_I, leading to the final result in equation (4.101.1). The concentration, $I(T)$, of iodine at any time T, before equilibrium is attained, is obtained from equation (2.189.3) as

$$I(T) = \frac{\gamma_I \Sigma_f \phi}{\lambda_I}(1 - e^{-\lambda_I T}) + I(0)e^{-\lambda_I T},$$

where $I(0)$ is the initial concentration of iodine-135. In the case where T is reckoned from the time of reactor start-up, $I(0)$ is zero.

4.102. The rate of increase in the xenon concentration is given by an expression analogous to equation (2.191.1), i.e.,

$$\frac{dX}{dT} = -\lambda_X X - \sigma_X \phi X + \lambda_I I + \gamma_X \Sigma_X \phi,$$

where the symbol X is used to represent properties associated with xenon. The direct fission yield of xenon-135, represented by γ_X, is 0.003, as stated above. The equilibrium concentration, X_0, of xenon is obtained from equation (2.193.1) as

$$X_0 = \frac{\lambda_I I_0 + \gamma_X \Sigma_f \phi}{\lambda_X^*} = \frac{(\gamma_I + \gamma_X)\Sigma_f \phi}{\lambda_X^*}, \qquad (4.102.1)$$

where $\lambda_X^* \equiv \lambda_X + \sigma_X \phi$; because σ_X is so high, λ_X^* is appreciably greater than λ_X. At any time T after initial start-up, so that $I(0)$ and $X(0)$ are zero, the concentration of xenon is given by equation (2.192.4) as

$$X(T) = X_0(1 - e^{-\lambda_X^* T}) + \frac{\lambda_I I_0}{\lambda_X^* - \lambda_I}(e^{-\lambda_X^* T} - e^{-\lambda_I T}). \qquad (4.102.2)$$

4.103. The macroscopic absorption cross section of xenon at any time T is equal to $X(T)\sigma_X$, and so the corresponding poisoning, $P(T)$, defined by equation (4.97.1), becomes

$$P(T) = \frac{X(T)\sigma_X}{\Sigma_u}, \qquad (4.103.1)$$

where $X(T)$ is given by equation (4.102.2). The equilibrium value of the poisoning, P_0, is obtained by substituting X_0 for $X(T)$ in equation (4.103.1); hence, using equation (4.102.1) for X_0

$$P_0 = \frac{X_0 \sigma_X}{\Sigma_u} = \frac{\sigma_X(\gamma_I + \gamma_X)\Sigma_f \phi}{\lambda_X^* \Sigma_u}. \qquad (4.103.2)$$

4.104. For uranium-235, $\Sigma_f/\Sigma_u = \sigma_f/(\sigma_f + \sigma_c)$, where σ_f and σ_c are the fission and capture (nonfission) cross sections; these are seen, from Table 2.10, to be 549 and 101 barns, respectively, so that Σ_f/Σ_u is 0.84. As seen above, $\gamma_I + \gamma_X$ is 0.056 + 0.003, i.e., 0.059; σ_X is 3.5×10^5 barns, i.e., 3.5×10^{-18} cm^2; and λ_X, the radioactive decay constant of xenon-135, is 2.1×10^{-5} sec^{-1}. Equation (4.103.2) can thus be written as

$$P_0 = \frac{1.7 \times 10^{-19}\phi}{(2.1 \times 10^{-5}) + (3.5 \times 10^{-18}\phi)}, \qquad (4.104.1)$$

recalling that $\lambda_X^* = \lambda_X + \sigma_X \phi$. Some values of P_0, calculated from equation (4.104.1), for various steady-state fluxes, are given in Table 4.6 to two significant

figures. It is seen that the equilibrium poisoning is small for a thermal flux of 10^{12} neutrons/(cm^2)(sec), but increases rapidly at higher fluxes and approaches a limiting value.

TABLE 4.6. EQUILIBRIUM VALUES OF XENON POISONING DURING OPERATION OF REACTOR

Thermal Flux (ϕ)	*Poisoning* (P_0)
10^{12}	0.0070
10^{13}	0.030
10^{14}	0.046
10^{15}	0.048

4.105. If ϕ is about 10^{11} (or less) neutrons/(cm^2)(sec), the second term in the denominator of equation (4.104.1) may be neglected in comparison with the first; then

$$P_0 \approx 8 \times 10^{-15}\phi,$$

and the poisoning will be negligible, i.e., 8×10^{-4} or less. Even for a flux of 10^{12} neutrons/(cm^2)(sec), the poisoning is only 0.007, so that about 0.7 per cent of the thermal neutrons are absorbed by the equilibrium amount of xenon. However, for values of ϕ greater than 10^{12}, the poisoning increases rapidly, at first (Fig. 4.11). If the flux is 10^{15} (or more) neutrons/(cm^2)(sec), λ_X can be neglected in comparison with $\sigma_X \phi$, so that $\lambda_X^* \approx \sigma_X \phi$; the poisoning, as given by equation (4.103.2), then reaches a limiting value expressed by

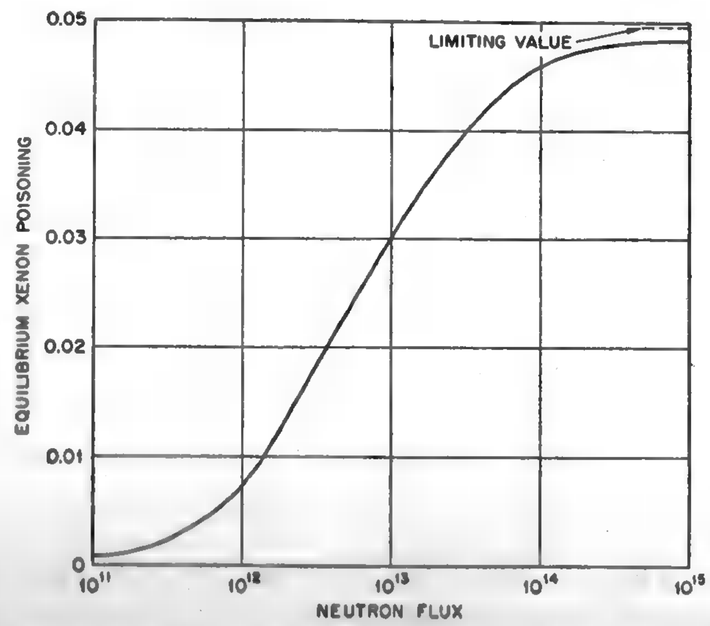


FIG. 4.11. Equilibrium values of xenon poisoning during reactor operation

$$P_{\text{lim}} \approx (\gamma_I + \gamma_X)\frac{\Sigma_f}{\Sigma_u} = (0.059)(0.084)$$
$$= 0.050.$$

This is the maximum value of the poisoning *during operation* for thermal reactors, with uranium-235 as fuel, no matter how high the flux. In view of the relationship between the poisoning and reactivity derived in § 4.99, it is seen that the maximum decrease in reactivity due to xenon poisoning during reactor operation is about 0.05. The maximum after shut-down can, however, be many times this value.

Xenon Poisoning After Shut-down

4.106. Because iodine-135 has a shorter half life than xenon-135, i.e., $\lambda_I > \lambda_X$, the conditions are suitable for the xenon concentration to increase to a maximum after the reactor is shut down completely. As explained in § 2.198, this occurs because the iodine-135 present at shut-down forms xenon-135 by radioactive decay at a rate that is initially greater than the rate of decay of the xenon-135, almost none of the latter now being lost by neutron capture. The concentration $X(t)$ of xenon at time t after shut-down is obtained from equation (2.197.1) as

$$X(t) = \frac{\lambda_I}{\lambda_X - \lambda_I} I_0(e^{-\lambda_I t} - e^{-\lambda_X t}) + X_0 e^{-\lambda_X t}, \qquad (4.106.1)$$

assuming that equilibrium has been attained before shut-down. The first term on the right gives the xenon concentration due to the decay of the iodine-135 followed by decay of the xenon-135 thus formed after shut-down; the second term, on the other hand, represents the decay of the xenon present in the reactor prior to shut-down. Since I_0 and X_0 depend on the neutron flux during operation, it is evident that this will also affect the xenon "build-up," as it is called, after shut-down.

4.107. In the derivation of equation (2.197.1), upon which equation (4.106.1) is based, it was assumed that the flux dropped to zero immediately when the reactor was shut down. Although this is not strictly true for a reactor, it is an adequate approximation for the purpose at hand. Taking into consideration the initial rapid drop and the subsequent stable negative period of 80 sec due to delayed neutrons (§ 4.41), it is found that a reactor can be shut down from a flux of 10^{12} to 10^{8} neutrons/(cm²)(sec) in about 20 min. This time is short compared with the several hours during which the xenon concentration builds up, and so very little error results from the assumption that the neutron flux drops immediately to zero when the reactor is shut down.

4.108. From equation (4.103.1), which expresses the poisoning, and equation (4.106.1) for the xenon concentration, it is possible to determine $P(t)$ at various times after shut-down for different values of the neutron flux before shut-down. The results for thermal fluxes of 10^{13}, 10^{14}, and 2×10^{14} neutrons/(cm²)(sec) are indicated by the curves in Fig. 4.12. For fluxes of the order of 10^{13} (or less)

neutrons/(cm²)(sec), the increase in poisoning after shut-down is seen to be negligible, but it assumes greater significance for higher operating fluxes. Thus, for a flux of 2×10^{14} neutrons/(cm²)(sec), the poisoning maximum, attained about 11 hr after shut-down,* is 0.51; this may be compared with the equilibrium value of less than 0.05 when the reactor was in operation. When the flux is 10^{15} neutrons/(cm²)(sec), the maximum xenon poisoning attained after shut-down is

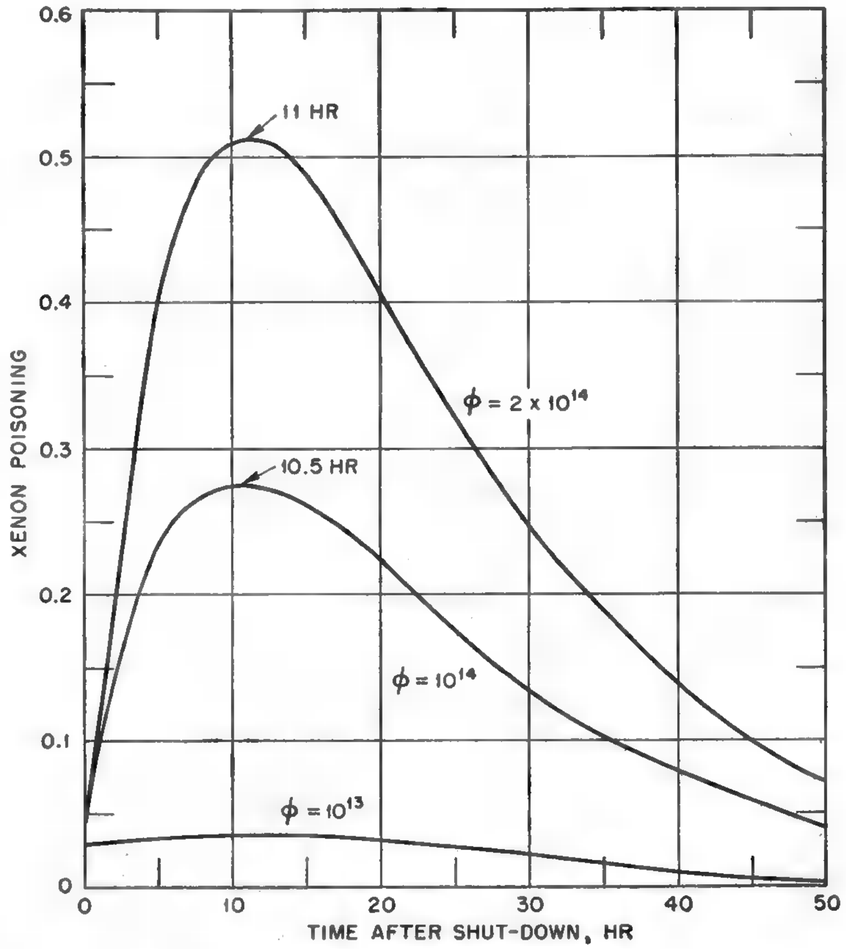


Fig. 4.12. Change of xenon poisoning after shut-down

roughly 2.5; the value is so large as to make it a practical impossibility to operate a thermal reactor at such a high steady-state flux. An operating flux of about 2×10^{14} neutrons/(cm²)(sec) would be as high as seems reasonable, unless a procedure is developed for continuous removal of the progenitors of xenon-135, especially the iodine-135, while the reactor is in operation.

* The time after shut-down at which the maximum concentration is reached can be calculated from equation (2.199.1).

Example A thermal reactor, using uranium-235 as fissionable material, has been operating for some time at a flux of 2×10^{14} neutrons/(cm²)(sec); how long after shut-down will the xenon poisoning reach a maximum and what is the poisoning at this time?

The time to attain the maximum xenon-135 concentration (or poisoning) after shut-down is obtained from equation (2.199.1), where A represents iodine-135 and B is xenon-135. The expressions for A_0 and B_0 are consequently given by equations (4.101.1) and (4.102.1), respectively. The value of λ_A, the radioactive decay constant for iodine-135, is 2.9×10^{-5} sec^{-1} and λ_B for xenon-135 is 2.1×10^{-5} sec^{-1}. The absorption cross section of iodine-135 is negligible, but σ_B, for xenon-135, has been given above as 3.5×10^{-18} cm² for thermal neutrons. The fission yields, as already stated, are 0.056 for iodine-135 and 0.003 for xenon-135. Since the equation for t_{max} involves the ratio of B_0 to A_0, the value of Σ_f is not required. Upon inserting the foregoing data into equation (2.199.1), the time to attain maximum xenon poisoning after shut-down is found to be 3.95×10^4 sec, i.e., close to 11 hr.

If this value for t is inserted into equation (4.106.1) for $X(t)$, the xenon concentration after shut-down, and the result is combined with the general equation (4.103.1) for the xenon poisoning, the maximum value of the latter can be calculated. The only additional information required is Σ_f/Σ_u, and this is known to be 0.84 for uranium-235. As a result the maximum value of the xenon poisoning is estimated to be about 0.51. (It will be noted that the contribution of the last term on the right of equation (4.106.1) is small; this is due to the fact that the xenon present after shut-down is due almost entirely to the decay of the iodine-135.)

4.109. In view of the approximate equality of reactivity decrease and the poisoning, it is evident that if a high-flux reactor is to be capable of being restarted at any time after shut-down, it must have an appreciable amount of excess multiplication (or reactivity), in the form of additional fuel, built into it. In order that a thermal reactor, normally operating at a flux of 2×10^{14} neutrons/(cm²)(sec), shall be able to "override" the maximum xenon concentration after shut-down, it must have about 0.51 of excess reactivity available.

4.110. As an alternative to the capability of overriding the xenon poisoning at any time, a reactor may be designed with sufficient reactivity available to override the xenon within a certain limited time only, e.g., 30 min, after shut-down. For a flux of 2×10^{14} neutrons/(cm²)(sec), this is considerably less than the maximum of 0.51 referred to above. However, should the delay in restarting be greater than the specified time, it may be necessary to wait a day or more before the reactor can be started up again. As soon as the flux becomes appreciable, after start-up, the xenon concentration will decrease rapidly toward the equilibrium value for the particular steady-state flux.

4.111. One way in which an excessive rise in the xenon concentration can be prevented is for the flux to be decreased to such an extent on shut-down that, although the power output is a fraction only of its operating value, the flux is still quite substantial. The removal of xenon-135, as a result of neutron capture, would thus continue after shut-down even though at a reduced rate, and this would prevent the poisoning from becoming too serious. In accordance with the arguments in §§ 2.196, 2.197, it is found that if ϕ' is the neutron flux after shut-down, then

$$X(t) = e^{-\lambda_X' t}\left[\int_0^t (\lambda_I I + \gamma_X \Sigma_f \phi')e^{\lambda_X' t}\, dt + X_0\right], \qquad (4.111.1)$$

where

$$I = \frac{\gamma_I \Sigma_f \phi'}{\lambda_I}(1 - e^{-\lambda_I t}) + I_0 e^{-\lambda_I t}$$

and

$$\lambda_X' = \lambda_X + \sigma_X \phi'.$$

The equilibrium concentrations I_0 and X_0 refer to those attained before shut-down when the flux was ϕ [cf. equations (4.101.1) and (4.102.1)]. If the reactor has not been operating long enough for the iodine and xenon equilibria to be established, I_0 and X_0 must be replaced by the actual concentrations at the time of shut-down. By using the appropriate expression for ϕ', equation (4.111.1) may be solved for various types of shut-down, e.g., partial instantaneous shut-down, partial or complete shut-down in steps, or linear (or other) partial or complete shut-down.

4.112. Although the differential equations for the concentrations of iodine and xenon attained under various conditions can frequently be solved analytically, considerable time can be saved, especially in the more complicated cases, by the use of an electrical simulator.* Because the time periods required for noticeable changes are of the order of hours, an electromechanical device is preferable to the resistance-capacitance type of circuit described earlier. Such a system can be designed as a true simulator, with the same time scale as an actual reactor. In view of the long time involved, however, it is preferable to change the time scale, so that the simulated processes of growth and decay of xenon can be speeded up by a large, known factor.

4.113. It should be noted that the equations given above have involved the assumption of uniform distribution of neutron flux, as well as of iodine-135 and xenon-135, throughout the reactor. If ϕ is taken as the average flux, the resulting error in the value of the poisoning will be no more than a few per cent. A more exact treatment is possible, using perturbation theory, but the method used here will suffice for all preliminary purposes, at least.

Samarium Poisoning

4.114. Next to xenon-135, the most important fission product poison is samarium-149. This is a stable isotope with an absorption cross section of 5.3×10^4 barns for thermal neutrons. It is the end product of the decay chain

$$Nd^{149} \xrightarrow{1.7h} Pm^{149} \xrightarrow{47h} Sm^{149}\ \text{(stable)},$$

which occurs in about 1.4 per cent of uranium-235 fissions by slow neutrons. Since the half life (1.7 hr) of neodymium-149 is short in comparison with that of promethium-149 (47 hr), it may be supposed that the latter is a direct product

* For references, see ‖ 4.02.

of fission, its fractional yield being 0.014. The problem of the formation of promethium and of samarium in a reactor is thus analogous to that considered above for iodine and xenon, respectively. As before, the absorption cross section of promethium-149 is so small that $\sigma\phi$ can be neglected in comparison with λ. Because the samarium-149 is not radioactive, it is found that the equilibrium concentration and the poisoning during reactor operation are independent of the neutron flux.

4.115. Taking γ for promethium as 0.014 and for samarium as zero, the values of λ as 4.1×10^{-6} sec^{-1} and zero, respectively, since samarium-149 is a stable isotope, and σ for samarium-149 as 5.3×10^{-23} cm^2, the equilibrium value of the poisoning is calculated to be 0.012. The maximum change in reactivity due to samarium in an operating reactor is thus -0.012, irrespective of the thermal-neutron flux.

4.116. After shut-down, the samarium concentration does not pass through a maximum, as does that of xenon, but it increases toward an asymptotic value (P_∞) represented by

$$P_\infty = \frac{\Sigma_f}{\Sigma_u}\left(\frac{\sigma_S \phi \gamma_P}{\lambda_P} + \gamma_P\right),$$

where the subscripts P and S stand for promethium and samarium, respectively; ϕ is the steady-state flux prior to instantaneous shut-down. Inserting the known values for Σ_f, Σ_u, σ_S, γ_P, and λ_P, it is found that

$$P_\infty = 1.5 \times 10^{-16}\phi + 0.012.$$

Thus, for a steady-state flux of 2×10^{14} neutrons/(cm^2)(sec), which appears to be the practical limit for a thermal reactor, the samarium poisoning after instantaneous shut-down increases to 0.042. The presence of samarium-149 in a reactor thus requires, at most, an addition of 0.04 to the reactivity, so that samarium poisoning is a minor problem compared with that of xenon poisoning.

Symbols Used in Chapter IV

$A_0, A_1, \ldots$	constants
B^2	buckling
$B_0, B_1, \ldots$	constants
C	capacitance
C	precursor nuclei/cm^3 (weighted average)
C_i	precursor nuclei/cm^3 (ith kind)
C_{i0}	C_i at $t = 0$
C_i'	small change in C_i
C_{ia}	complex amplitude of C_i'
D	diffusion coefficient
d	density
E	voltage
E_i	input voltage

E_o	output voltage
e	base of natural logarithms
f	thermal utilization
f'	thermal utilization with poison
I	current
I	iodine concentration (nuclei/cm^3)
I_0	iodine concentration at equilibrium
$I(0)$	iodine concentration at $T = 0$
$I(T)$	iodine concentration at time T
Ih	inhour unit
k	infinite multiplication factor
k_{eff}	effective multiplication factor
k'_{eff}	k_{eff} with poison
k_0	k_{eff} at $t = 0$
k'	small change in k_{eff}
k_a	complex amplitude of k'
k_{ex}	excess multiplication factor
L	inductance
L	diffusion length (also at temperature T)
L_0	diffusion length at temperature T_0
L_m	diffusion length in moderator
l	neutron lifetime in finite medium
l_0	neutron lifetime in infinite medium
M	migration length (also at temperature T)
M_0	migration length at temperature T_0
m	number of delayed neutron groups
n	neutron density (neutrons/cm^3)
n_0	neutron density at $t = 0$
n'	small change in neutron density
n_a	complex amplitude of n'
P	poisoning (Σ_p/Σ_u)
P_0	equilibrium poisoning (during operation)
$P(T)$	poisoning at time T (during operation)
P_{lim}	limiting value of P_0
P_∞	asymptotic value of samarium poisoning
p	resonance escape probability
R, R_0	resistances
R	radius of spherical reactor at temperature T
R_0	radius of spherical reactor at temperature T_0
r	rate of change of multiplication factor
$\mathbf{r}$	general spatial coordinate
S	neutron source strength
S_0	constant (primary) source strength

T	time of reactor operation
T	reactor period (stable)
T_0	initial reactor period
T, T_0	absolute temperatures
$T(t)$	time-variable component of neutron flux
t	time
t	time after shut-down
t_{max}	time for maximum poisoning after shut-down
$t_{\frac{1}{2}}$	radioactive (or delayed neutron) half life
V	volume at temperature T
V_0	volume at temperature T_0
v	velocity
X	xenon concentration (nuclei/cm^3)
X_0	xenon concentration at equilibrium
$X(T)$	xenon concentration at time T (during operation)
$X(t)$	xenon concentration at time t (after shut-down)
z	ratio Σ_u/Σ_m
α	coefficient of linear expansion
β	total fraction of delayed fission neutrons
β_i	fraction of delayed fission neutrons of ith kind
γ	fission yield
γ_I	fission yield of iodine-135
γ_P	fission yield of promethium-149
γ_X	fission yield of xenon-135
λ	radioactive decay constant
λ	precursor decay constant (weighted average)
λ_i	precursor decay constant (ith kind)
λ^*	effective decay constant ($\lambda + \sigma\phi$)
λ_I	decay constant of iodine-135
λ_P	decay constant of promethium-149
λ_X	decay constant of xenon-135
ρ	reactivity
Σ_a	absorption cross section (macroscopic)
Σ_f	fission cross section (macroscopic)
Σ_m	absorption cross section of moderator (macroscopic)
Σ_p	absorption cross section of poison (macroscopic)
Σ_s	scattering cross section (macroscopic)
Σ_u	absorption cross section of fuel (macroscopic)
σ_a	absorption cross section at temperature T
σ_{a0}	absorption cross section at temperature T_0
σ_I	absorption cross section of iodine-135
σ_P	absorption cross section of promethium-149
σ_X	absorption cross section of xenon-135

τ	Fermi age (also at temperature T)
τ_0	Fermi age at temperature T_0
ϕ	neutron flux
ϕ_0	neutron flux at $t = 0$
ϕ'	neutron flux after shut-down
$\phi(\mathbf{r}, t)$	neutron flux at point $\mathbf{r}$ at time t
$\phi(\mathbf{r})$	spatial component of neutron flux
φ	phase angle
ω	operator (d/dt)
ω	parameter (reciprocal time)
$\omega_0, \omega_1, \ldots$	values of parameter ω, solutions of equation
ω_0	reciprocal of stable period
ω_f	oscillation frequency
∇^2	Laplacian operator

PROBLEMS

1. Using the data in Table 2.11, determine the weighted average decay constant for the delayed fission neutrons from (*a*) uranium-233, (*b*) plutonium-239.

2. From the results of the preceding problem, compare qualitatively the effect of a given change in the effective multiplication factor on the neutron flux in thermal reactors using uranium-233 or plutonium-239 as fissionable material with those employing uranium-235.

3. Derive equation (4.17.2) from equations (4.10.1) and (4.14.1) by using ω to represent the operator d/dt.

4. Assuming one average group of delayed neutrons, plot the variation of the thermal neutron flux with time (for the first 2 sec) after a natural-uranium reactor undergoes a sudden increase of 0.0015 in reactivity. The thermal-neutron lifetime in the reactor is 10^{-3} sec.

5. For the conditions of the preceding problem, estimate the time required for the transient contribution to the neutron flux to decrease to about 1 per cent of the total. Determine (*a*) the initial and (*b*) the stable value of the reactor period.

6. From the data in Table 4.1 and equation (4.35.1) plot a curve relating the reactivity to the reciprocal of the stable reactor period for a reactor in which the thermal-neutron lifetime is 10^{-3} sec; k_{eff} may be taken as unity. (Determine ρ for T values of 1, 5, 10, 25, and 50 sec and plot ρ versus $1/T$.) At what period does the magnitude of the neutron lifetime become significant?

7. Compare the results of taking into account all five groups of delayed neutrons, as in the preceding problem, by plotting ρ against $1/T$ for the case of one average group of delayed neutrons.

8. Repeat the preceding problem assuming the delayed neutrons to fall into two groups, one consisting of the two with the shortest half lives and the other of the three longer half lives (R. A. Fayram and K. Bernstein, *Chem. Eng. Prog. Symposium Series*, No. 12, **50**, 244 (1954)). What general conclusions can be drawn from the results of Problems 6, 7, and 8?

9. Verify the statement in § 4.115 that the equilibrium poisoning in an operating thermal reactor due to samarium-149 is about 0.012, independent of the neutron flux.

10. Derive an expression for the samarium-149 concentration in a reactor after shut-down and show that it attains the asymptotic value given in § 4.116.

11. After operating at a thermal flux of 10^{14} neutrons/(cm²)(sec) for a long time, a reactor is shut down completely. How long will it take the xenon concentration to rise to a maximum? What is the value of the xenon poisoning at that time?

12. Repeat the preceding problem assuming a thermal flux of 10^{15} neutrons/(cm²)(sec). From these results and those in the text, draw general conclusions as to the effect of the neutron flux on (*a*) the time required to attain the maximum xenon poisoning after shut-down, and (*b*) the value of the maximum poisoning.

13. Calculate the xenon poisoning for a reactor operating at a thermal flux of 10^{14} neutrons/(cm²)(sec) at the following times after initial start-up: (*a*) 20, (*b*) 30, and (*c*) 50 hr; and at the following times after complete shut-down: (*a*) 5, (*b*) 15, (*c*) 25, (*d*) 35, and (*e*) 50 hr. Combine the results with those from Problem 11 and plot a curve showing the variation of xenon poisoning, for the given flux, with time during 50 hr of operation followed by 50 hr of shut-down.

14. A homogeneous (Water Boiler type) reactor contains uranium-235 and water in the weight ratio of 1 to 15 (see Chapter III, Problem 15). Make a rough estimate of its temperature coefficient of reactivity, based on the approximation of a "large" reactor. The coefficient of volume expansion of water may be taken as 10^{-4} per °F.

*Chapter V**

NUCLEAR REACTOR INSTRUMENTATION

INTRODUCTION

5.1. The operation of a reactor involves a great variety of instrumentation, much of which is of a conventional character familiar to engineers. In this category, for example, are flow meters and thermocouples for determining the flow rates and temperatures, respectively, of the coolant. A knowledge of the distribution of coolant velocity and temperature throughout the core is important, especially in a reactor operating at fairly high power. Since the measuring devices used for these purposes, as well as those which indicate the positions or motions of control rods, do not present any unusual features they will not be considered in this chapter.

5.2. The average power output of a reactor can be obtained from the product of the temperature rise of the coolant and its total rate of flow; in fact, an electrical instrument, described in § 5.128, has been developed which indicates the product directly. However, for control purposes such indicators are not adequate since they do not respond instantaneously either to over-all or to local changes in the reactor power. As seen in § 2.163, the power level is proportional to the neutron flux, and instruments that measure flux can be located at many points throughout a reactor system. Such devices have an essentially instantaneous response and are particularly suitable for indicating power levels and for operating automatic control and safety mechanisms.

5.3. In addition to determinations of neutron flux, other measurements of nuclear radiations, e.g., of alpha and beta particles and of gamma rays, are performed in connection with reactor operation. For example, observations are made on the radioactivity of the coolant, such as air or water, emerging from the reactor to indicate possible leakage of fission products and to determine whether it can be disposed of safely. Radiation measuring instruments of various kinds are also required for experiments and tests associated with the design and operation of nuclear reactors. Finally, as part of the health physics program for

* Material contributed by S. Glasstone, W. H. Jordan, and J. D. Trimmer. Reviewed by W. H. Jordan.

preventing overexposure to radiations, regular surveys are made with suitable instruments of both operating areas and personnel.

5.4. No matter what their purpose, most devices for the detection and measurement of nuclear radiations are based on the same fundamental principles and these will be considered here. However, consideration of the actual instrumentation will be restricted to that employed in reactor control and operation, including determination of power output and its variation during start-up and shut-down. The instruments and devices used in personnel protection (monitoring) activities will be described in Chapter IX, in connection with the general treatment of radiation protection.

NUCLEAR RADIATIONS AND IONIZATION

Direct and Indirect Ionization

5.5. The property of nuclear radiations, including neutrons, most widely used for their measurement is their ability to cause ionization, either directly or indirectly, of the medium, e.g., a gas, through which they pass. The problem of measuring nuclear radiations is reduced, therefore, to devising methods for the observation of the resulting ionization. As seen in Chapter II, alpha and beta particles form ion-pairs directly by interaction with atoms or molecules. Although neutrons and gamma rays cannot produce ionization directly, they can be made to do so by indirect means.

5.6. Each of the three major types of interaction of gamma rays with matter, i.e., the Compton effect, photoelectric effect, and pair production (see § 2.47, *et seq.*), results in the liberation of free (recoil) electrons. The energy of these electrons depends on the circumstances, but it is usually high enough for them to be capable of producing considerable ionization. Thus, the indirect production of ion-pairs due to the passage of gamma rays through a medium occurs more or less automatically, i.e., without any special means being necessary.

5.7. With neutrons the situation is somewhat different, since specific nuclear interactions are required to produce ionizing particles. Three main types of reaction have been used: (1) elastic scattering by hydrogen nuclei; (2) the (n, α) reaction with boron; and (3) fission. The first of these processes is applicable only to fast neutrons, of energy about 0.1 Mev or more; in their passage through hydrogen gas, or a compound rich in hydrogen, e.g., a hydrocarbon, the fast neutrons can transfer a large proportion of their energy to hydrogen nuclei (§ 3.55). The resulting recoil protons, moving with high speed, produce considerable ionization in their passage through a gas.

5.8. The second and third reactions referred to above are used for the detection of slow neutrons. The reaction of such neutrons with boron-10 produces a helium nucleus (alpha particle) and a lithium nucleus (§ 2.92), both of which

have relatively high energy and are capable of causing ionization.* Slow neutrons are able to induce fission of uranium-235 and the resulting fission fragments can produce large numbers of ion-pairs in their paths (§ 2.176).

5.9. The (n, α) reaction with boron-10 and fission of uranium-235 have hitherto played the major role in neutron detection, but there are two other processes to which reference should be made because of their possible future importance. One of these is the (n, α) reaction with lithium-6, described in § 2.93; this process has been applied to slow-neutron detection based on the scintillations produced by the resulting charged particles (see § 5.87, *et seq.*). The other is the (n, p) reaction of slow neutrons with helium-3 (§ 2.94); the ionization caused by the products can be readily observed.

5.10. Although not strictly in the same category as the processes described above whereby the neutrons are made to produce ion-pairs indirectly, mention may be made of another, even more indirect, means frequently used for neutron measurements. A material, such as indium, is exposed to slow neutrons and as a result of the (n, γ) reaction a radioactive isotope is formed (§ 2.84). The radiations, usually beta particles and gamma rays, emitted by the product are then detected by the ionization they cause (§ 5.102).

Behavior of Ion-pairs in Electric Field

5.11. Many instruments for the detection of nuclear radiations are dependent upon the behavior in an electric field of the ion-pairs formed by the ionizing particles in their passage through a gas. This behavior varies with the potential gradient of the electric field and can best be explained by considering two electrodes, e.g., a metal cylinder as one and a wire running along its axis as the other (Fig. 5.1), placed in a vessel of gas and a potential difference established across them. As a general rule, the central wire is the positive electrode (anode) and the outer cylinder the negative electrode (cathode), so that negative ions, usually electrons, are attracted to the former and positive ions to the latter.

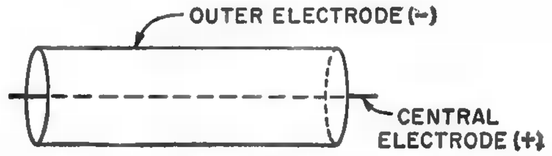


Fig. 5.1. Electrical field for study of ion-pairs

5.12. Suppose a burst (or pulse) of ionizing radiation enters the gas between the electrodes so that a definite number of ion-pairs are formed. Under the influence of the electric field the positive ions will move toward the negatively charged electrode and the negative ions will migrate toward the positive electrode. The magnitude of the charge collected on the electrodes will depend on the

* The Li^7 nucleus is usually in an excited state; the kinetic energy of the alpha particle is then 1.5 Mev (range 0.7 cm in air) and that of the lithium nucleus is 0.85 Mev (range 0.4 cm in air).

applied voltage and the general nature of the variation is indicated in Fig. 5.2, in which the charge collected is shown (on a logarithmic scale) as a function of the voltage. Curve A refers to a case in which the radiation pulse produces 10 ion-pairs and curve B to a larger pulse producing 1000 ion-pairs. It can be seen that the curves can be divided into six fairly distinct regions, marked I to VI. Three of these regions, namely, II, III, and V, are used in various types of nuclear radiation instruments, and these will be considered in turn.*

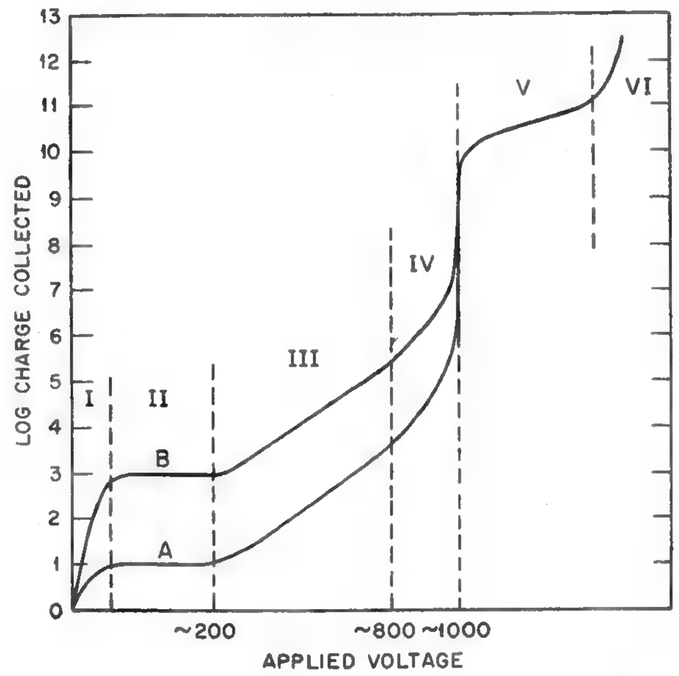


FIG. 5.2. Variation of charge collected with applied voltage

THE IONIZATION CHAMBER REGION

5.13. At very low voltages the charge collected at first increases with the applied voltage (region I) and then attains a constant value (region II). At very low potentials, the ions move slowly in the electrical field, with the result that many of them recombine before they reach the electrodes. With increasing voltage, the ions migrate more rapidly toward the respective electrodes; the extent of recombination is thus decreased and the charge collected on the electrodes increases. This accounts for the behavior in region I.

* D. H. Wilkinson, "Ionization Chambers and Counters," Cambridge University Press, 1950; B. B. Rossi and H. H. Staub, "Ionization Chambers and Counters: Experimental Techniques," National Nuclear Energy Series, Div. V, Vol. 2, McGraw-Hill Book Co., 1949; E. Segrè (Ed.), "Experimental Nuclear Physics," Vol. I, John Wiley and Sons, Inc., 1953 (Part I, "Detection Methods," by H. H. Staub); S. A. Korff, "Electron and Nuclear Counters," 2nd ed., D. Van Nostrand Co., Inc., 1955. For general review, see D. Taylor and J. Sharpe, *Proc. Inst. Elec. Eng.* (*Lond.*), **98**, Part II, 174, 209 (1951).

5.14. Above a certain voltage, the ions move so fast that the recombination is virtually zero, and essentially every ion produced by the nuclear radiation reaches the electrode. A further increase in the applied voltage will not result in any increase in the number of ion-pairs collected. Hence, for region II, referred to as the *ionization chamber* (or *ion chamber*) region, the charge remains constant as the voltage is increased. The charge collected is then equal to the total charge carried by the ion-pairs produced in the gas (ionization chamber) by the nuclear radiation. This is 10 unit charges (log 10 = 1) for case A and 1000 unit charges (log 1000 = 3) for case B, in Fig. 5.2.

THE PROPORTIONAL REGION: GAS AMPLIFICATION

5.15. The ionization chamber region has a limited range, and as the potential between the electrodes is increased, a new phenomenon appears. In region III, the quantity of charge collected for a given amount of radiation is larger than, but proportional to, the original amount of ionization; it has consequently been called the *proportional region.* The potential gradient is now so high that the (primary) electrons, i.e., the negatively charged members of the ion-pairs, move fast enough to be themselves capable of causing secondary ionization in the gas. The secondary electrons so produced may cause further ionization and so on. A single ion-pair can thus initiate a chain or "avalanche" of ionizations. The charge collected on the electrodes in the proportional region is therefore larger than that carried by the primary ion-pairs formed by the passage of radiation through the gas.

5.16. The total number of ion-pairs produced by a single, primary ion-pair is called the *gas-amplification factor*. This factor is unity in the ionization chamber region (II), but it may become as large as 10^4 or more in the proportional region (III). The charge collected in both of these regions is equal to the original number of ion-pairs multiplied by the amplification factor. Consequently in Fig. 5.2 the curves A and B remain a constant distance apart, because of the logarithmic scale of the ordinates, in regions II and III.

5.17. It will be apparent from the figure that in the proportional region the gas-amplification factor increases rapidly, in a roughly exponential manner, with the applied voltage. For any given voltage, however, it has a definite value, depending upon the arrangement of the electrodes and the nature of the gas in which the ionization occurs.

5.18. As the voltage is increased other factors arise which limit the production of secondary ion-pairs, so that the increase in the gas-amplification factor does not continue indefinitely. The atmosphere of positive ions around the central (positive) electrode gives rise to a positive space charge which causes a decrease in the electric field, thus tending to decrease the extent of gas amplification. At sufficiently high voltage, e.g., in region IV, the amplification factor consequently approaches a limit and the charge collected is no longer proportional to

the initial ionization. Region IV is therefore referred to as the limited proportional region.

The Geiger Region

5.19. In region V, known as the *Geiger region*, the ionization avalanche, which has hitherto been confined to a point or small region of the (positive) electrode, spreads over the whole of its length. The fact that the curves A and B in Fig. 5.2 now coincide means that the total amount of charge collected is independent of the extent of the primary ionization, i.e., of the number of ion-pairs produced by the pulse of nuclear radiation. Thus, although the gas-amplification factor is very large, it does not have a definite value, as it does in the proportional region. The charge collected increases with the applied voltage, but its amount does not depend on the nature or energy of the radiation causing the initial ionization. In the Geiger region (V), therefore, an ionizing particle giving rise to a large number of primary ion-pairs will cause essentially the same amount of charge to be collected on the electrodes as will a particle producing a small number.

5.20. The characteristics of the Geiger region can be considered from another point of view. Suppose a source of ionizing radiations is placed near the vessel containing the gas and the electrodes, as described above. The electrical circuit is such that each particle entering the gas can produce a pulse of ionization capable of detection in a suitable manner, as will be seen below. The detecting device (or counter) is able to record only relatively large pulses, but not small ones.

5.21. Until the applied voltage reaches a certain value, referred to as the *starting potential*, the pulses are too small to be detected. With increasing voltage the gas amplification increases and the pulses get larger and larger, but may vary in size, depending upon the initial ionization. Eventually, at the *Geiger threshold*, corresponding to the beginning of region V in Fig. 5.2, the pulses are all the same size. As the voltage is increased further, the pulses become large enough to be recorded by the counting device. If the counting rate is plotted against the voltage between the electrodes, a curve similar to that in Fig. 5.3 is obtained. It is apparent that over a certain range of potential, called the *Geiger plateau*, the rate at which ionization pulses are recorded is essentially constant.

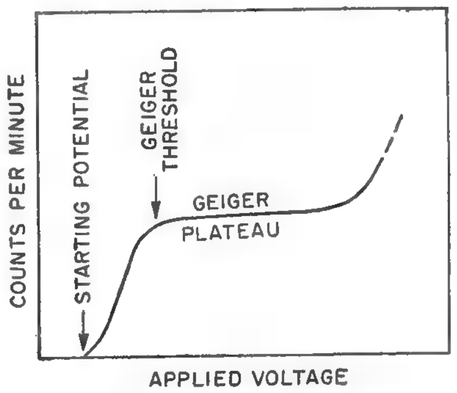


Fig. 5.3. Pulse counting-rate as function of applied voltage

5.22. Almost every particle entering the chamber, whether it causes little or much primary ionization, is now recorded because in the Geiger region the total ionization produced is virtually independent of the initial effect. Although the Geiger plateau is represented as horizontal, this is an ideal situation. In actual practice there is usually a slight upward slope with increasing potential, but by suitable design it can be made quite small, so that in the Geiger region the rate at which pulses are recorded from a given source may be regarded as being constant and independent of the applied voltage.

5.23. If the applied voltage is increased beyond the Geiger region, there is a rapid increase in the charge collected; this represents region VI of Fig. 5.2. The potential is then so high that, once a discharge is initiated, others follow in rapid succession, so that there is effectively a continuous discharge. This region is not used for the detection or measurement of ionizing radiations.

IONIZATION CHAMBERS*

Integrating Instruments: Electroscopes

5.24. A number of different instruments for the measurement of nuclear radiations operate in the ionization chamber region, i.e., where the charge collected on the electrodes is equal to that carried by the primary ion-pairs. Although there are marked differences even among the instruments in each category, ionization-chamber measurements fall into two types, namely, integrating and nonintegrating. In the integrating instruments, which will be considered first, the total quantity of charge due to a number of ionizing particles is collected over a period of time. In the nonintegrating (or counting) devices, on the other hand, each particle capable of causing ionization is recorded separately.

5.25. Integrating devices also can be divided into two classes, which may be referred to as electrostatic and electrodynamic (or current indicating), respectively. Those of the electrostatic type are often called *electroscopes*, since they operate on the same principle as the familiar gold-leaf electroscope, which was used in some of the earliest measurements of radioactivity. In one form of this instrument two thin sheets of gold are given an electric charge, so that they repel one another. As long as no ions are present, the leaves will retain their charge and will remain in the same position. If, however, ions are formed, e.g., by nuclear radiations, those of the appropriate charge will be attracted to the gold leaves. This will cause a reduction in the charge and a decrease in the mutual repulsion of the leaves, which will thus gradually come together again.

5.26. Electroscopes of various types, operating on the same general principles as described above, are being used in the nuclear energy field especially in connection with radiation surveys. For this purpose they are designed to be sensitive, rugged, and portable. The Lauritsen electroscope, for example, which will be described in Chapter IX, is used for measuring gamma radiations as part of area surveying (monitoring). Because of their penetrating power these radiations readily pass through the walls of the ionization chamber. If

* See references to § 5.12.

required for the detection of beta (or alpha) particles, the radiations are allowed to enter the chamber through a "window" of thin aluminum or other light material. With the window "open" the instrument will measure both beta and gamma radiations, but if it is closed gamma rays only will be detected.

5.27. Simple forms of integrating ionization chambers of the electrostatic type, not much larger than a fountain pen in size, are used extensively in health physics work to determine the total amount of radiation (or dosage) to which an individual has been exposed over a period of time. These devices will also be considered more fully in the chapter dealing with radiation protection.

Integrating Chambers with Current Measurement

5.28. In the second type of integrating ionization chamber, a constant potential is maintained between the electrodes by means of a battery (Fig. 5.4). If ionizing radiation enters the chamber at a sufficiently high rate, the ions produced are swept continuously to the respective electrodes, and a steady current

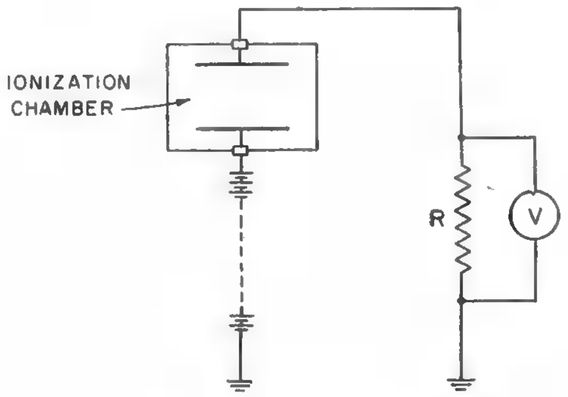


Fig. 5.4. Integrating type of ion-chamber circuit

flows. The strength of this ion current is a direct measure of the rate of entry of the ionizing particles and hence of the radiation intensity. The ion current may be measured directly, or it may be determined by means of a high-impedance voltmeter V connected across a resistance R through which the current flows. The reading on the voltmeter is then proportional to the ion current and consequently to the radiation intensity.

5.29. Ionization chambers with current-measuring devices have been designed for many different purposes. In the parallel-plate chamber, as its name implies, the electrodes are two parallel plates, which may be flat or curved (§ 5.39). Some chambers have central electrodes consisting of a single wire or a network of wires, insulated from the container, e.g., a cylindrical vessel, which serves as the other electrode. The electrode to which the measuring instrument is attached is called the *collecting electrode*. This is frequently maintained at (or close to) ground potential, while the other (high-voltage) electrode is kept at

the required voltage to permit operation in the ionization-chamber region. The gas filling the chamber may be air, carbon dioxide, nitrogen, argon, or methane, among others.

5.30. Because of their adaptability, integrating ionization-chamber instruments are widely used in nuclear energy work, both for radiation monitoring and for other purposes. Like the electroscopes, they can be made to indicate gamma-radiation intensities only, or by means of suitable windows they can be used to determine alpha or beta particles (or both) in addition. They have been adapted also to the measurement of fast or slow neutrons.

Neutron Detection

5.31. For the detection of fast neutrons the ionization chamber is filled with hydrogen or with a hydrocarbon gas; alternatively, or in addition, the interior walls may be lined with a hydrogenous substance, such as paraffin or polyethylene. If exposed to fast neutrons, recoil protons of high energy will be liberated, as indicated above. The ion current due to the ionization produced by the recoil protons is then a measure of the neutron density.

5.32. In monitoring operations it is frequently necessary to measure the neutron intensity in a fairly high field of gamma radiation. The arrangement just described, however, would indicate both fast neutrons and gamma rays. In order to determine the neutrons alone, a differential system has been devised. This consists of two similar ionization chambers; one contains hydrogenous material, so that it is sensitive to both fast neutrons and gamma radiation, whereas the other is filled with argon or other gas in which the gamma rays produce almost all of the (indirect) ionization. The pressures of the gases in the chambers are adjusted so that when exposed to the same gamma radiation they give equal ion currents. In a region containing both neutrons and gamma rays, the difference in the ion currents will then give a measure of the fast-neutron density. One serious drawback of this type of differential system is that it is unsatisfactory for use with radiation beams that do not subtend both chambers equally.

5.33. In principle, the adaptation of an ionization chamber to the measurement of slow neutrons is a simple matter. All that is necessary is to coat one or both electrodes with boron, preferably enriched in the boron-10 isotope, since this is the one that has a high cross section for the reaction with slow neutrons. Alternatively, the chamber may be filled with gaseous boron trifluoride or with a gas enriched in helium-3 (§ 5.9). Integrating ionization chambers for neutron detection have also been made by coating the electrodes with a uranium-235 compound.

Ionization Chambers for Reactor Control

5.34. For use in reactor control there are certain special requirements which introduce difficulties. The chamber must be small, since many are needed in a

reactor, yet it must deliver a reasonably large ion current in order to provide rapid and smooth response. These two conditions mean that the chamber must be placed in a region of high neutron flux. As a result, problems of radiation damage (see Chapter VIII), particularly to the insulators, become acute. Furthermore, the chamber materials become highly radioactive; this not only means that repair is difficult but, more serious, the beta particles emitted by these materials interfere with the neutron measurement. In some reactors, placing the chamber in a high neutron flux means that it is subjected to high temperatures, and this further complicates the problem of construction. A great deal of research has consequently gone into the question of chamber design, and much still remains to be done.

5.35. In order to obtain as large as possible an ion current, there are certain optimum values for the thickness of the boron coating and the spacing of the electrodes. By using enriched boron, it appears that the best that can be expected is a current of 10^{-16} amp/cm^2 of boron-coated electrode surface per unit of flux. Thus, if a chamber with an active area of 100 cm^2 is placed in a neutron flux of 10^{10} neutrons/(cm^2)(sec), the ion current will be roughly 10^{-4} amp. This is about as large a current as can conveniently be drawn from an ionization chamber of this size while still preserving the proportionality between ion current and neutron flux.

5.36. If the chamber is to operate in regions of relatively low flux, the coated electrode area should be made as large as possible. In some instruments, the electrodes are parallel and cup-shaped, thus providing a relatively large area within a small volume.* The same objective has been achieved in an ionization chamber using a number of flat, circular parallel (boron-coated) plate electrodes, connected alternately and arranged perpendicular to the direction of neutron travel (Fig. 5.5).† This instrument, called the PCP (parallel circular-plate) chamber, was designed to respond quickly to a change in the neutron flux in the reactor core; it delivers a current of 10^{-5} amp in a streaming thermal flux of

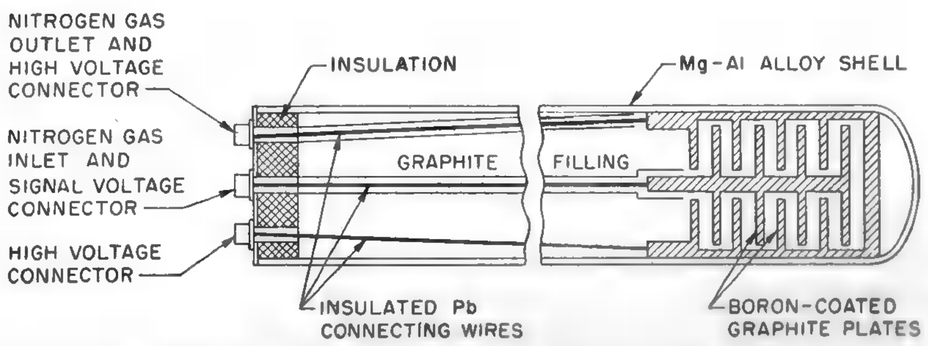


FIG. 5.5. Parallel circular-plate (PCP) ionization chamber

* H. Carmichael, unpublished; see also § 5.39.
† R. K. Abele and J. C. Gundlach, Report AECD-3494.

2.5×10^9 neutrons/(cm^2)(sec). By keeping the chamber in a region where the flux is relatively low (cf. § 5.110), and by using construction materials, e.g., high-purity graphite for the electrodes and a magnesium-aluminum alloy shell, which do not become very radioactive under neutron bombardment, the chamber can be handled safely 30 min after the reactor is shut down.

Differential and Compensated Chambers

5.37. Both for control and monitoring it is frequently necessary to determine the slow-neutron flux in the presence of fairly intense gamma radiation. One way in which this can be achieved is by the use of two chambers, as described in § 5.32. One chamber contains boron trifluoride gas and is sensitive to both slow neutrons and gamma rays; the other contains argon and responds only to the gamma radiation. The difference in the ion current is then a measure of the slow-neutron flux. This differential method can be used for monitoring, but it is not satisfactory for reactor control, because the device is too cumbersome for the purpose.

5.38. The principle of the compensated ionization chamber designed for reactor control purposes may be explained with the aid of Fig. 5.6. There are three electrodes A, B, and C; of these, B is the collecting electrode, which is grounded, and A and C are the accelerating electrodes, whose potentials are

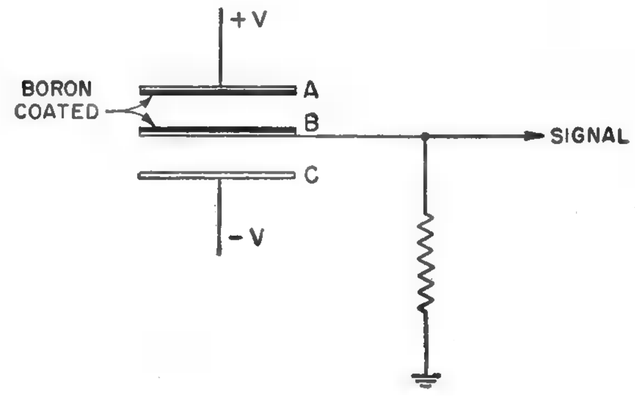


FIG. 5.6. Compensated ion chamber for neutron detection

opposite with respect to ground. If the position of B is such that the volumes AB and BC are roughly equal, the amounts of ionization produced in them by gamma rays will be approximately the same. The net ion current to the collecting electrode will then be almost zero. If now boron is coated on the surfaces of one volume of the chamber, e.g., AB, the net ion current will be a measure of the neutron flux.

5.39. A drawing of the gamma-compensated ionization chamber, developed at Oak Ridge National Laboratory for use in reactor control, is given in Fig. 5.7. The electrodes consist of three graphite cups which fit together. Provision

is made for adjusting the compensating volume; the best position of the inner cup depends on the gradient of the gamma-ray flux and must be determined for the particular location in the reactor. The outside diameter of the chamber is $3\frac{3}{4}$ in. and its total length is about 36 in., although the sensitive region is restricted to a few inches at the end. The compensated ion current may vary from 10^{-4} amp, at full operating level of the reactor, down to 10^{-10} amp, and even to 10^{-12} amp, so that the instrument can be used over at least a 10^6-fold power (or neutron flux) range.

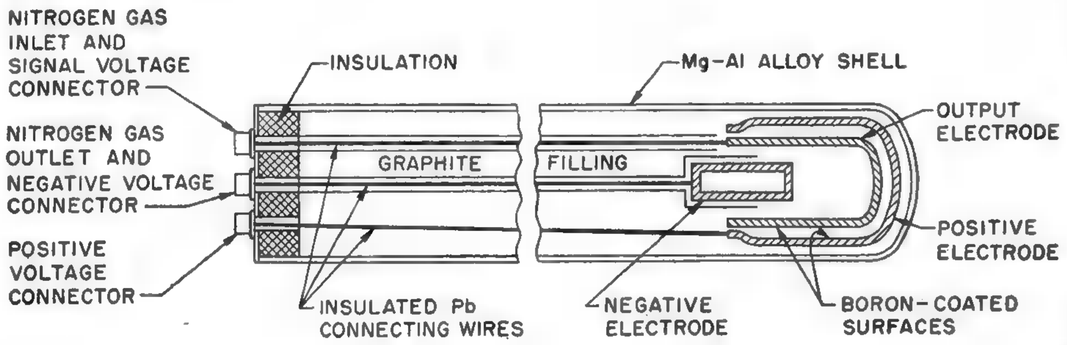


FIG. 5.7. Gamma-compensated ionization chamber

DEVICES FOR MEASUREMENT OF ION CURRENT

MAGNITUDE AND RANGE OF CURRENT

5.40. One of the main problems associated with the use of ionization chambers is the measurement of the small ion currents. Suppose an instrument of this type, with boron-coated electrodes, is being used for the detection of neutrons; as seen above, the energy of the alpha particle liberated for each neutron reacting is 1.5 Mev. Its range in air is about 0.7 cm, and so if the plates of the chamber are at least this distance apart, it may be supposed that the whole of the 1.5-Mev energy is expended in causing ionization in the chamber. It requires roughly 33 ev to form one ion-pair in air, and so the alpha particle will produce 4.5×10^4 ion-pairs. The charge carried by each ion of a pair is, in general, equal to the electronic charge, i.e., 1.6×10^{-19} coulomb (amp-sec). The alpha particles liberated in the chamber at the rate of 1 per sec will thus produce an ion current of $(4.5 \times 10^4)(1.6 \times 10^{-19}) = 7.2 \times 10^{-15}$ amp. The 0.85-Mev lithium ion will produce an ion current of somewhat more than half this amount. Hence the total ion current may be assumed to be approximately 10^{-14} amp per neutron per sec reacting with boron in the ionization chamber.

5.41. The neutron flux in a reactor will vary over a considerable range, from shut-down to full operating level, and the rate of alpha-particle and lithium-ion production in an ionization chamber may easily range from 10^4 (or less) to 10^{10} (or more) per sec. The corresponding range of the ion current would thus be from about 10^{-10} to 10^{-4} amp; these appear to be roughly the practical limits for satisfactory operation of an ionization chamber. For the measurement of currents less than 10^{-10} amp or so, long integration times are required and then statistical fluctuations make the results unreliable (§ 5.97, *et seq.*).

5.42. Many devices have been developed which are suitable for measuring the currents produced by ionization chambers. No one instrument can be said to be superior to others for all purposes; generally, one particular method of measurement will be best suited to each type of application, involving certain current magnitudes and ranges. Some of the chief methods for determining ion currents, and the conditions under which they are employed, will be considered in the following sections.

GALVANOMETERS

5.43. Currents in the range of 10^{-6} to 10^{-4} amp, i.e., 1 to 100 microamp, can be readily indicated on a microammeter. In spite of their simplicity, such instruments are of limited applicability, since more is often required than mere indication of the ion current on a short scale. However, when combined with a magnetic relay, such as the Sensitrol,* microammeters can be used to operate safety circuits, e.g., when the neutron flux exceeds a predetermined value (see Chapter VI).

5.44. The suspension type galvanometer has been employed in the past in reactor work; in conjunction with a neutron-sensitive chamber, it is sometimes the principal instrument used for indicating the power level of a reactor. A single galvanometer with shunts can be used to measure currents with considerable precision over the effective range of a good ionization chamber, e.g., 10^{-10} to 10^{-4} amp. The sensitivity of the galvanometer can be increased by inclusion of a potentiometer which supplies a compensating current, leaving only the difference current to be read on the scale of the instrument. For example, suppose the ion current at operating level of the reactor is the full-scale reading of 100 microamp. If now a constant compensating current of 99 microamp is supplied to the galvanometer, the sensitivity can be increased to 1 microamp for full-scale. A change of 0.1 per cent in the power of the reactor would then correspond to a 10 per cent change in the galvanometer reading.

5.45. Galvanometers have been found useful in the operation of a reactor at widely different power levels for experimental purposes. An important advantage is the fact that they do not employ electronic tubes, thus eliminating a possible source of failure. Their chief disadvantages are their slow response to changes in current and the fairly elaborate installation required to make them insensitive to mechanical vibrations.

* It may be noted that there has been some question concerning the reliability of so light a relay.

Vibrating-Capacitor Electrometer

5.46. Vibrating-capacitor electrometers,* sometimes called vibrating-reed electrometers, are finding increasing application for the measurement of small currents. They are used, in conjunction with ionization chambers, not so much for indicating the neutron flux (or power) in reactor control, but rather in subsidiary aspects, such as monitoring the radioactivity of the coolant, e.g., air or water, after passing through the reactor. Vibrating-capacitor electrometers are useful in such applications because they will measure currents as low as 10^{-12} amp with a reasonably short response time. They can be used for currents down to 10^{-17} amp, although with proportionately longer response times. The instruments can operate for long periods without requiring recalibration or resetting the zero.

5.47. The principle of operation of the vibrating-capacitor electrometer may be explained by reference to Fig. 5.8; current from an ionization chamber flows first through an isolating resistor R_0 and then through the resistance R_1, thereby generating a voltage (V) at A, and a corresponding charge ($Q = CV$) on the capacitor C. By means of an electromagnetic vibrator, the capacitance of C is varied so rapidly, usually 60 cps, that the charge does not have time to become

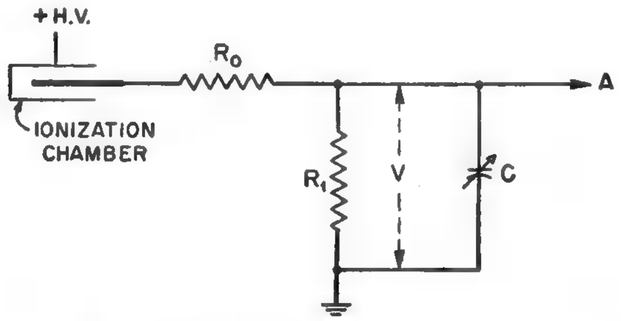


Fig. 5.8. Vibrating-capacitor electrometer circuit

redistributed during one cycle. As a result, the voltage at A is modulated at the capacitor vibration frequency. The modulated voltage can then be amplified by means of an a-c amplifier and the rectified output voltage used to operate a meter, a relay, or a recorder, as required, according to the purpose of the instrument. If a recorder is to be used, it may prove more economical to employ a combined electrometer-recorder,† since many commercial recorders are already equipped with a-c amplifiers and phase-sensitive detectors.

* H. Palevsky, R. K. Swank, and R. Grenchik, *Rev. Sci. Inst.*, **18**, 298 (1947).
† H. Reese, Jr., *Nucleonics*, **6**, No. 3, 41 (1950).

D-C Amplifiers

5.48. Direct-current amplifiers have been applied for many years for measuring small currents.* While they can be used for currents down to 10^{-17} amp, reactor operation seldom involves indication of currents below 10^{-12} amp. Although much simpler than vibrating-reed electrometers, they are more subject to drift. A typical requirement for a d-c amplifier would be somewhat as follows. A neutron-sensitive ionization chamber delivers an ion current of 10 microamp at normal reactor power, and it is desired that this small current should produce a 30-volt signal with a low impedance for operating a safety circuit. In addition, the neutron flux (or power level) as measured by the ion current, should be indicated on a 10-millivolt recorder. A possible scheme for accomplishing these results is shown in Fig. 5.9.

5.49. The vacuum tube is operated as a cathode follower† with a gain of slightly less than unity. If R_1 is chosen as 3.3 megohms, a signal of 33 volts will be developed across it in the case postulated above. This will produce a signal at the cathode of approximately 30 volts, plus the grid-to-cathode bias

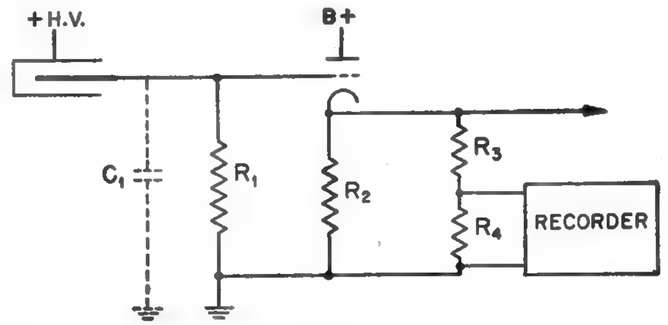


Fig. 5.9. Recording of neutron flux by ion chamber and d-c amplifier

which can be canceled if necessary. A resistor divider network (R_3, R_4) is then used to provide the 10 millivolts to the recorder. Since the signal from the tube is large, drifts due to changes in contact potentials (which may be as high as 0.1 volt) will not affect the signal voltage appreciably. It is an important, but sometimes overlooked, condition that the signal voltage at the grid of the tube must be much larger than the contact potential drifts.

5.50. The method described can be used for much smaller currents; it is only necessary to increase R_1 in order to maintain a large signal voltage at the grid of the tube. There are, however, two factors which place a limit upon going to smaller and smaller currents. One is the grid current in the tube, i.e., the cur-

* For circuit details, see W. C. Elmore and M. L. Sands, "Electronics: Experimental Techniques," National Nuclear Energy Series, Div. V, Vol. 1, McGraw-Hill Book Co., 1949, p. 180, *et seq.*; K. Kandiah and D. E. Brown, *Proc. Inst. Elec. Eng.* (*Lond.*), **99**, Part II, 314 (1952).
† Cf. W. C. Elmore and M. L. Sands, *op. cit.*, p. 56.

rent collected on the grid due to electrons and positive ions produced in the tube itself. This limitation can be overcome by the use of special electrometer tubes, such as the General Electric FP-54, with grid currents of less than 10^{-14} amp, which are well below the ion currents usually encountered in reactor measurements.

5.51. The second factor setting a lower limit on the current which can be measured by means of a d-c amplifier is associated with the time constant (RC) of the input circuit. In this case, it is equal to the product of the resistance R_1, in parallel with C_1 (Fig. 5.9), and the capacitance of the ionization chamber and connecting cables. As R_1 is increased, in order to measure smaller ion currents, the time constant becomes very long and the meter responds very slowly. Since long cables are generally necessary with ionization chambers used for reactor control, the problem is particularly serious. For example, if R_1 is 10^{12} ohms and C_1 is 500$\mu\mu$f, the time constant is 500 sec; this would be much too long for reactor control, since rapid response is vital.

5.52. One solution to the problem is to design ionization chambers to give larger currents, and this is done whenever possible. An alternative is to use electronic circuits that produce the required output voltage of about 30 volts with signal voltages of a few millivolts, so that R_1 can be kept reasonably small. This involves the use of d-c amplifiers to amplify the signal voltage, with the attendant difficulties due to drift in contact potentials. Changes in the gain of the amplifier due to tube aging can usually be minimized by feedback networks. If neither of the foregoing measures proves altogether satisfactory, then the vibrating-capacitor electrometer, described in the preceding section, will be a better, although more expensive, solution to the problem of measuring very small currents.

Logarithmic Amplifiers

5.53. Because of the wide range in neutron flux encountered during start-up of a reactor, it is convenient to have a meter that reads the ion current on a logarithmic scale. This can be achieved essentially by substituting a thermionic diode for the resistance R_1 in Fig. 5.9. Within certain current limits, e.g., about 10^{-4} to 10^{-12} amp, the voltage across such a diode is proportional to the logarithm of the current flowing through it. An instrument of this kind is called a "log n meter," since it indicates the logarithm of the neutron density (or flux).

5.54. The general outline of the circuit for a logarithmic amplifier is indicated in Fig. 5.10.* The current from the ionization chamber is led directly to the plate of a thermionic diode, which provides the only path to ground. When the ion current is very small, the plate of the diode assumes a negative potential of a few volts. As the current increases, the plate voltage, V, increases toward

* W. H. Jordan, H. B. Frey, and G. Kelley, Report ORNL-110, C. W. Johnstone, Report AECU-363; J. Weill, *Nucleonics*, 11, No. 3, 36 (1953).

zero according to the expression $V = B \ln ki$, where i is the ion current and B and k are constants. The voltage change across the diode is only about 0.2 volt per 10-fold change in the ion current, and so a voltage amplifier is required. Thus, one with a gain of 100 will produce approximately 20 volts output for a 10-fold change in the neutron flux. The input resistance of the amplifier must be high if all the chamber ion-current is to flow through the diode. The output terminals can be connected directly to a strip chart recorder.

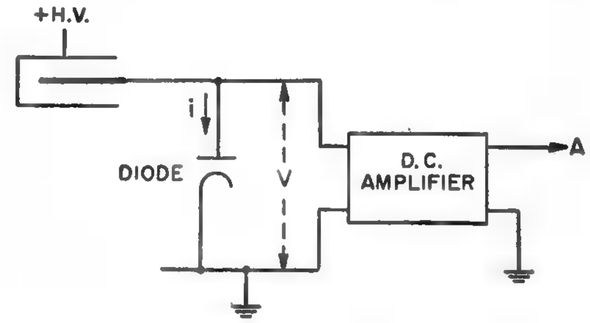


Fig. 5.10. Logarithmic amplifier circuit

5.55. One of the disadvantages of the logarithmic amplifier is that it cannot be relied upon for accurate measurement of neutron flux, because of drifts due to changes in the diode. The situation can be improved somewhat by means of balanced diodes and differential amplifiers. However, the main use of the logarithmic amplifier is to indicate the reactor period (§ 4.9) during start-up or shut-down; in this connection the drifts are not a serious handicap.

Period Meters

5.56. For the direct determination of reactor period the output of the logarithmic amplifier is fed into an RC differentiating circuit, as in Fig. 5.11. As seen above, the voltage at A is given by

$$V = B \ln ki, \qquad (5.56.1)$$

where i is the ion current which is proportional to the neutron flux (or density). If i_0 is the ion current at time $t = 0$, and i is the value at time t, then it follows from equation (4.9.1) that

$$i = i_0 e^{t/T}, \qquad (5.56.2)$$

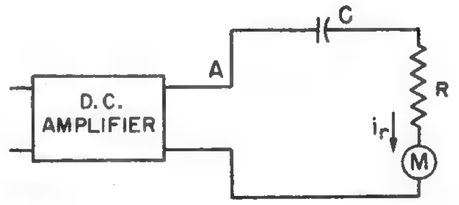


Fig. 5.11. Diagram of differentiating circuit

where T is the stable (or constant) reactor period. Upon combining equations (5.56.1) and (5.56.2) the result is

$$V = V_0 + B\frac{t}{T}, \qquad (5.56.3)$$

where V_0 is the initial voltage. The current i_r flowing through the meter M is equal to $C(dV/dt)$ plus a transient term which decays exponentially with a time constant RC. Upon substituting the value of dV/dt obtained from equation (5.56.3), it is found that

$$i_r = \frac{CB}{T},$$

so that the current i_r read on the meter M is inversely proportional to the reactor period.

5.57. The period meter M may be a microammeter with a scale calibrated in seconds, so that the period can be read directly. A meter with an offset zero can be used to indicate both positive and negative periods. Since the voltage drop across R, or a portion of it, is also a measure of the current i_r, it can be used to operate a recorder or to provide a signal for activating a safety circuit.

5.58. The tracings obtained on recorder charts attached to a logarithmic amplifier (log n meter) and to a period meter during shut-down and start-up of a reactor are reproduced in Figs. 5.12 and 5.13, respectively. It should be noted that the time scales run from right to left in each case, and that in the

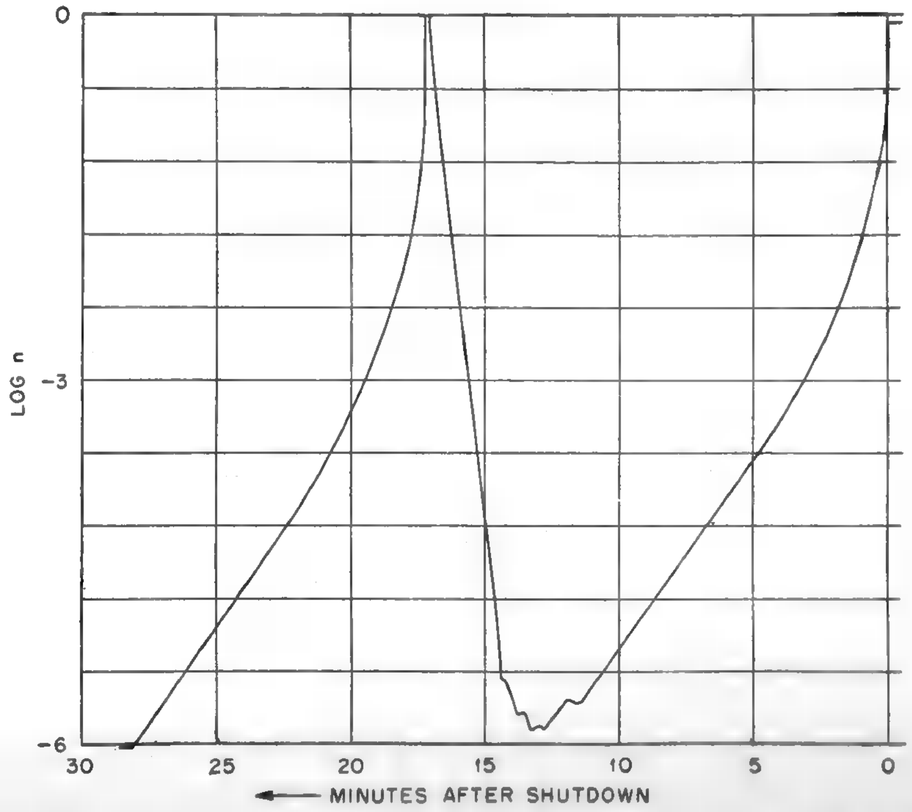


FIG. 5.12. Recorder tracing of log n accompanying insertion and withdrawal of control rods

latter figure the record is in two parts (upper and lower), because the chart speed was faster.

5.59. At time $t = 0$, the control rods were inserted so as to shut down the reactor (Fig. 5.12); the neutron density dropped, at first very rapidly, and then more slowly because of the effect of the delayed neutrons (§ 4.40). At time $t = 12$ min, one control rod was withdrawn, causing a transient rise in the neutron density. However, the reactor was still subcritical, and so the transient rise was followed by a further decrease. The transient changes accompanying the withdrawal of other control rods are seen on the chart, until at $t = 14$ min, the reactor became supercritical and the flux began to increase exponentially, as

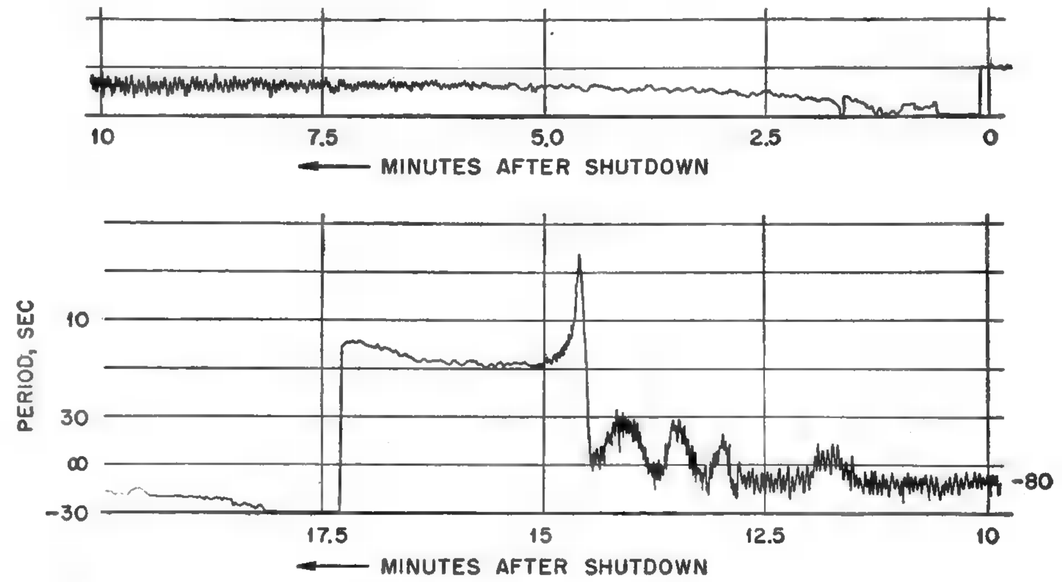


FIG. 5.13. Tracing of period indicator based on differentiation of log n curve

indicated by the straight line on the logarithmic chart. At $t = 17$ min a safety circuit tripped, because the neutron flux was too high, and the reactor was automatically shut down.

5.60. The corresponding tracing on the period meter which gives, in effect, the slope of the logarithmic curve in Fig. 5.12, is shown in Fig. 5.13. Between $t = 5$ and 12 min, the line indicating the (negative) reactor period is approximately horizontal. Although there is considerable fluctuation or "noise" (§ 5.100), it can be seen that the period was roughly constant, at about −80 sec, corresponding to the linear portion of Fig. 5.12, between 5 and 12 min. The transients due to withdrawal of the control rods are seen from $t = 12$ to 14 min. Between $t = 15$ and 17 min, the (positive) period was approximately constant at 15 sec; then, when the safety rods dropped, at $t = 17$ min, because the maximum allowable power was exceeded, the period became negative.

NONINTEGRATING IONIZATION CHAMBERS*

Pulse Chambers

5.61. The foregoing discussion has referred to ionization chambers of the integrating type; some consideration will now be given to nonintegrating devices, or *particle counters*, which record individual ionizing events. The basic difference between an integrating system and a particle counter lies in the values of their RC time constants relative to the frequency of arrival of the particles. If an ionization chamber is to function as a particle counter, the time required to indicate the voltage change resulting from collection on the electrodes of the ions produced by a single particle must be considerably less than the average time between the arrival of successive particles. Consequently a particle counter circuit must have a relatively short time constant; the ionization burst due to the entry of each separate particle can then be detected. In instruments of the integrating type, the time constant is long; the ion current may then be regarded as a continuous succession of ionization bursts which cannot be recorded individually because of the slow response of the system.

5.62. The schematic arrangement of an ionization-chamber circuit for pulse counting is shown in Fig. 5.14. The collecting electrode is connected to a suitable amplifier, so that the pulses are amplified (and modified) before going on to activate a counting instrument, as will be described below. The time constant of the chamber is determined by the resistance R and the associated capacitance C. If RC of the detecting system is fairly large, then there will be sufficient time for recording the slow-moving positive ions formed in each ionization pulse. The system is then called an *ion-pulse chamber;* the pulse height is proportional to the total number of ion-pairs produced in the chamber, irrespective of the position where they are formed. The main disadvantage of the ion-pulse system is that the relatively large time constant, e.g., 0.01 sec, makes it unsuitable for fast counting. Such chambers are not generally employed in reactor operation.

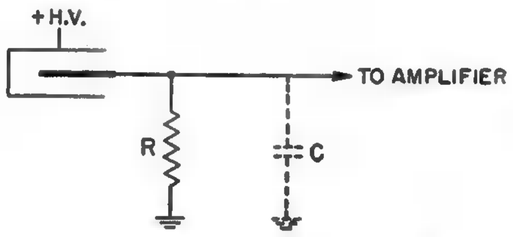


Fig. 5.14. Ionization-chamber circuit for pulse counting

5.63. If the over-all time constant is decreased to about 20 microsec or less, by differentiation in the amplifier, the positive ions are not recorded and the system operates on electron collection only; there is then obtained an *electron-pulse chamber*. Such a chamber, which requires that the negative parts of the ion-pairs shall exist mainly as free electrons, can be used to record pulses following one another at short time intervals. This means that the *resolving time*, i.e., the time elapsing between pulses which can be recorded separately, can be quite small. However, unless suitably modified by means of a grid between the electrodes, the size of the pulse is not independent of the position in the chamber where the ionization is produced.

* D. R. Corson and R. R. Wilson, *Rev. Sci. Inst.*, **19**, 207 (1948); R. R. Wilson, D. R. Corson, and C. P. Baker, "Particle and Quantum Detectors," National Research Council, 1950; S. C. Curran and J. D. Craggs, "Counting Tubes: Theory and Application," Academic Press, Inc., 1949; see also references to § 5.12.

5.64. The chief problem in the operation of an electron-pulse type ionization chamber is maintaining the purity of the filling gas. In order to produce a quick change in the potential of the electrode, required for counting, it is necessary that the electrons formed by the entry of an ionizing particle should remain free and thus be capable of rapid migration to the positive electrode. If they become attached to gas molecules, to form negative molecular (or atomic) ions, their rate of movement is greatly decreased. Gases such as hydrogen, nitrogen, and the noble gases have a very small affinity for electrons and so, when pure, are suitable as filling gases for pulse counters. Traces of oxygen, water vapor, or any halogen must be avoided, because they readily capture electrons. Consequently if a sealed chamber is to be used, it must be thoroughly baked and out-gassed before filling, and it must be completely airtight. In many cases fresh gas is flowed continuously through the chamber; tank argon is satisfactory for this purpose.

5.65. One of the important advantages of the pulse chamber is that it can be used to determine strongly ionizing particles, e.g., alpha particles, directly and neutrons indirectly, in the presence of considerable intensities of radiation having a low specific ionization, e.g., beta particles and gamma rays. If the ionization chamber is fairly small, e.g., a few centimeters between the electrodes, then the number of ion-pairs produced by an alpha particle or, indirectly, by a neutron will greatly exceed that due to entry of a beta particle or gamma-ray photon. The voltage pulses will, accordingly, be much greater with the strongly ionizing particles. It will be seen below that the output pulses from the chamber are fed into an amplifier followed by a scaler or counting-rate meter; if the latter is properly biased, it will respond only to the large pulses and not at all to the weak ones. This discrimination might fail if the alpha particle or neutron density were very low while that of the gamma radiation, for example, is high. The weak pulses due to the latter might arrive at such frequent intervals that they would "pile up" on one another to produce, effectively, one large pulse to which the counter (or scaler) would respond. Such a situation can sometimes be met by decreasing the time constant (see § 5.69).

Pulse Chambers for Neutron Detection

5.66. For detecting neutrons, the pulse chamber may be lined with boron or filled with boron trifluoride. Most generally, however, the interior of the chamber (or the electrode surface) is coated with a uranium compound enriched with uranium-235; it is then called a *fission chamber*. The entry of a slow neutron

causes fission of the uranium, and the resulting fragments produce considerable ionization in the gas. Fission chambers have been found particularly useful for measuring neutron flux during the early stages of reactor start-up and shut-down. The pulse due to the fission fragments is so large that there is no difficulty in discriminating even against "pile-up" pulses from gamma rays. This is, of course, very important soon after shut-down, when the gamma-ray intensity is considerable.

5.67. A pulse-type fission chamber designed for reactor operation is shown in Fig. 5.15, together with some associated equipment.* The chamber itself is approximately 4 in. long and 1 in. in diameter. A nickel cylinder coated on the inside with uranium-235 (approximately 1 mg/cm^2) fits snugly inside the chamber. The central (collecting) electrode is an aluminum cylinder, of $\frac{3}{16}$ in. diam-

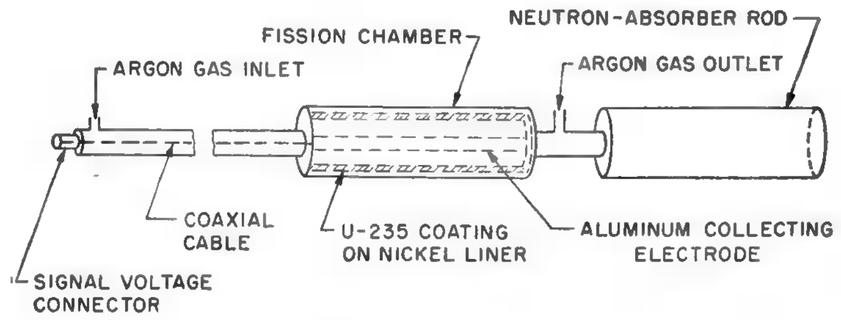


FIG. 5.15. Pulse-type fission chamber

eter, supported by the coaxial connector. Provision is made for attaching a neutron-absorber rod to the end of the chamber, so that withdrawal of the chamber and absorber into a cylindrical hole in the concrete shield will greatly reduce the neutron flux to which the chamber is exposed.

PULSE SHAPING AND AMPLIFICATION

5.68. Since there is no internal (gas) amplification in the ionization chamber, the relatively small voltage changes due to ionization bursts must be amplified in order to actuate a counter. In addition, the shape of the output pulse is such as not to lend itself to rapid counting, and so it must be modified in an appropriate manner. The passage of an ionizing particle through the ionization chamber produces a voltage change on the (positive) collecting electrode of the form depicted in Fig. 5.16. There is at first a slow rise due to amplifier limitations, followed by a rapid rise as the electrons move toward the collecting electrode. This process is complete in about 1 microsec or less. Subsequently, the voltage decays slowly, because of leakage, at a rate dependent upon R and C in Fig. 5.14. The best signal-to-noise ratio is obtained by making R large, e.g.,

* Oak Ridge National Laboratory, unpublished.

10 to 100 megohms. The time constant of the chamber is then large and the pulse voltage may take some hundreds of microseconds to decay.

5.69. In order to obtain a pulse of short duration, suitable for counting, the original output pulse from the ionization chamber is first amplified, by means of a linear amplifier.* Here the pulse is increased 20- or 30-fold, and it then passes to a differentiating circuit (Fig. 5.11), which gives an output voltage pulse of the form of Fig. 5.17. This pulse is seen to rise and fall sharply, in a short time interval, so that it is suitable for fast counting. It is further amplified by a factor of 1000 or so to produce a pulse with a height of 10 to 100 volts. The duration of the pulse depends on the RC characteristics of the amplifier. The ORNL A-1 amplifier† has provision for a stepwise variation of the pulse length

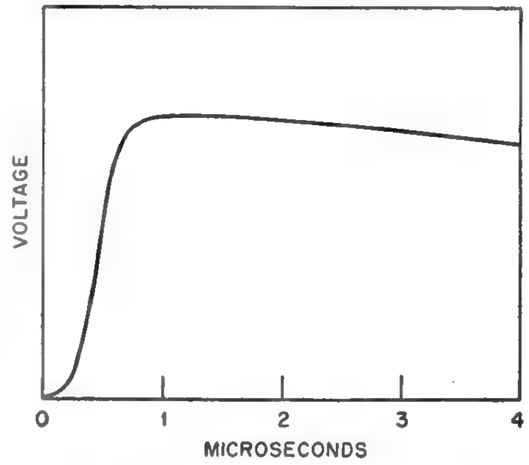


FIG. 5.16. Shape of voltage pulse from ionization chamber

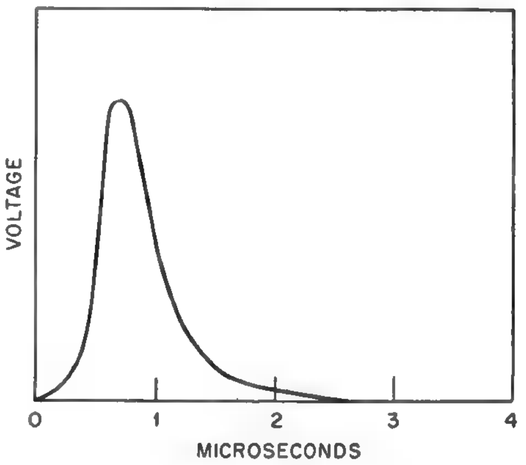


FIG. 5.17. Shape of voltage pulse after amplification and differentiation

from about 0.7 to 20 microsec. Smaller pulses can be detected in the "slow" position, but discrimination against the gamma-ray background, if present, can best be achieved in the "fast" position.

5.70. When amplified, as described above, the voltage pulse can actuate a mechanical counter, so that the number of pulses produced, and hence the number of particles entering the ionization chamber, can be recorded. As a general rule, mechanical recorders cannot operate fast enough, and so an electronic scaler is inserted between the amplifier and the recorder which permits a specified fraction only of the pulses to be counted. For many aspects of reactor operation it is necessary to obtain a rapid indication of the radiation, e.g., neutron, density. In such cases a counting-rate meter is used in conjunction with a pulse chamber and its associated amplifier. Since scalers and counting-rate meters are employed in connection with proportional and Geiger counters, as

* For a general description of voltage amplifiers, see W. C. Elmore and M. L. Sands, *op. cit.*, Chapter 3.

† W. H. Jordan and P. R. Bell, *Rev. Sci. Inst.*, **18**, 703 (1947).

well as with ionization chambers, for recording pulses, a more detailed description of these instruments will be given later (§ 5.91, *et seq.*).

PROPORTIONAL COUNTERS*

General Characteristics

5.71. The use of the proportional region III of Fig. 5.2 for the counting of ionizing particles has certain advantages. In the first place, the internal (gas) amplification means that less external amplification is required to make the voltage pulses large enough to actuate a mechanical counter or scaler. In the second place, a properly designed proportional counter has a very short recovery time, and so very high counting rates are possible. Since the extent of gas amplification increases with the voltage between the electrodes, it is possible, by operating at moderate voltages, to count highly ionizing particles in the presence of those having a low specific ionization. Then, by increasing the voltage, both kinds can be counted simultaneously, if desired.

5.72. One disadvantage of proportional counters, as compared with ionization chambers, is that the former require much higher voltages, e.g., 500 volts or more. Further, although variations in the applied voltage have little or no effect on the operation of an ionization chamber, such variations will alter the gas amplification, and so the output, of a proportional counter.

5.73. The electrodes in the proportional counter consist of a cylinder and a central wire, as in Fig. 5.1. The central wire is invariably made the anode (positive electrode) and this is usually the collecting electrode connected to the amplifier. The electric field between a cylinder and a central wire electrode is nonuniform, since it increases in proportion to $1/r$, where r is the distance from the wire. The field strength near the central electrode is therefore very large and a pulse of appreciable size, due to gas amplification, can result from the entry of an ionizing particle. In principle, the proportional region could be used in an integrating instrument in which the ion current is measured. But this would require such exact control of the electric field between the electrodes as not to be practical. Consequently pulse counting is always employed, and this accounts for the name "proportional counter."

Pulse Formation

5.74. The pulse from a proportional counter is formed in a different manner and differs in shape from that produced by an ionization chamber. The major role in pulse formation is now played by the motion of the positive ions, that of the electrons being relatively insignificant. Because of the high potential gradient near the central wire anode, most of the gas amplification occurs there. The positive ions formed are then accelerated rapidly outward, thus producing a voltage pulse with a sharp initial rise (Fig. 5.18). The subsequent gradual

* For references, see §§ 5.12, 5.61.

increase results from the slower motion of the positive ions in the regions of lower potential gradient a short distance away from the wire anode.

5.75. Since a pulse of the form of that in Fig. 5.18 is not of a good shape for counting, the output of the proportional counter is passed through a differentiating or "clipping" (RC) circuit. The exact shape of the pulse so obtained depends upon the time constant of the circuit, and two examples are shown in Fig. 5.19. It is seen that the use of an RC circuit with a short time constant gives a sharp output pulse suitable for rapid counting. The fact that the height of the pulse decreases as the time constant becomes shorter is not serious because the pulse can be amplified electronically, if necessary.

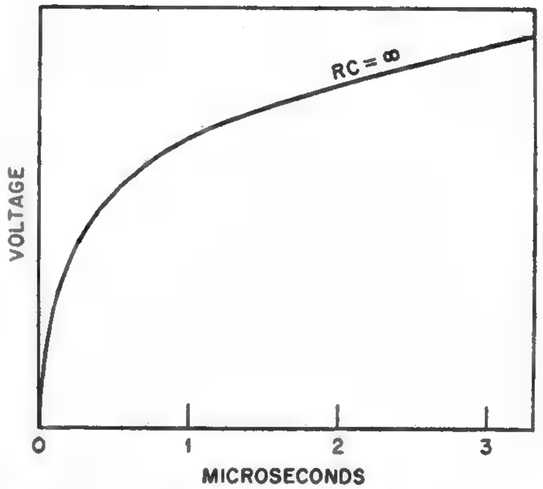


Fig. 5.18. Shape of voltage pulse from proportional counter

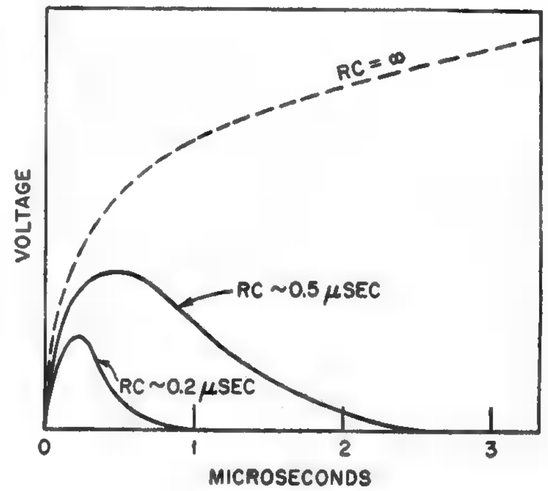


Fig. 5.19. Shape of voltage pulse after passage through a differentiating (or "clipping") circuit

5.76. When used in conjunction with a suitable clipping circuit with a small RC value, as described above, the resolving time of a proportional counter can be made very short, e.g., about 0.2 microsec or so. It should be pointed out, however, that the actual resolving time is not quite as short as would be implied by Fig. 5.19. The curve in Fig. 5.18 shows the voltage pulse increasing rapidly from zero time, but there is really a lag between the initial ionization and the rapid voltage rise. This is the time interval during which the primary electrons travel to the central wire anode. By using a suitable filling gas, e.g., a mixture of argon and carbon dioxide, the time lag can be reduced to about 0.1 microsec, so that the resolving time is not seriously affected.

Applications of Proportional Counters

5.77. As is the case with ionization chambers, proportional counters are particularly suited to the measurement of alpha particles and neutrons. They are

widely used for monitoring both personnel and areas for the presence of alpha-particle emitters (§ 9.97). For neutron counting the tube containing the electrodes is filled with boron trifluoride if slow neutrons are to be counted, and with hydrogen for the detection of fast neutrons. By applying the appropriate bias to the counting-rate meter or scaler, the weak pulses due to beta particles or gamma radiation can be eliminated. The small resolving time of the proportional counter makes discrimination possible even against "pile-up" pulses. In nuclear reactor work, proportional counters (containing boron trifluoride) are employed to some extent, although not where great accuracy is required (cf. § 5.115).

GEIGER-MÜLLER COUNTERS*

General Characteristics

5.78. Instruments operating in the Geiger region V of Fig. 5.2 are called Geiger-Müller counters (or Geiger counters), frequently abbreviated to G-M counters. Although the counter (or tube) itself can take various forms, both in size and shape, the electrodes almost invariably consist of a central wire anode, which is the collecting electrode, surrounded by a cylindrical cathode, just as for a proportional counter. The applied voltage is high, usually from 800 to 1500 volts, but its actual value is not critical, provided it is somewhere about the center of the Geiger plateau (§ 5.21). Thus the disadvantage of having to use such high voltages is partly offset by the fact that small variations will not affect the operation of the counter.

5.79. The main advantage of the G-M counter lies in the very considerable gas amplification, e.g., as high as 10^8, which is possible. The output pulses are often of the order of several volts, and little, if any, external amplification is required for the operation of a counter or counting-rate meter. However, reshaping of the voltage pulse is desirable.† The size of the voltage pulse from a G-M tube is essentially independent of the specific ionization (direct or indirect) caused by the nuclear radiation; thus, a neutron and a gamma-ray photon would produce the same effect. For this and other reasons, the G-M counter is not used directly in nuclear reactor operation. Its main function in the reactor field, apart from laboratory applications, is for monitoring relatively weak beta and gamma radiations.

5.80. The efficiency of a G-M counter for gamma-ray photons is quite small and varies with the energy. The efficiency can be improved, however, by utilizing the fact that, except for radiation of very low energy, pulses produced (indirectly) by photons are due to electrons resulting from the interaction of the gamma radiation with the walls of the G-M tube, rather than with the containing gas. By making the walls of a material of high atomic number, the probability (cross section) of interaction is increased and so also is the efficiency of the counter for photons. At the same time, the efficiency is made less sensitive to energy variations in the gamma radiations. For reasons given below, the resolving time of the G-M counter is, in general, relatively large compared with that of an ion chamber or proportional counter.

* See references in §§ 5.12, 5.61.

† R. P. Ghelardi and C. H. Brown, *Nucleonics*, 1, No. 1, 50 (1947).

Quenching the Discharge

5.81. A troublesome feature of the G-M tube is that, when an ionizing particle produces an avalanche of ion-pairs, the resulting discharge may itself initiate a series of further discharges. One particle may then give rise to several pulses or to a long continuous pulse. This phenomenon is attributed to the action of the positive ions. As a result of various direct and indirect processes, the impact of these ions upon the cathode is accompanied by the liberation of electrons. These are attracted to the central wire (anode) and in their path produce ionization which will cause a second discharge in the tube. In turn, this may be followed by another, and so on. Apart from the fact that these secondary discharges may give rise to spurious counts, the arrival of further primary pulses may go unrecorded.

5.82. There are two main methods for suppressing or *quenching* the discharge, so that it stops after each primary pulse. One method is by the use of an electrical quenching circuit which reduces the voltage below the starting potential (§ 5.21) momentarily after each pulse and restores it in time for the next pulse. Alternatively, a small quantity of a suitable organic vapor or a halogen (chlorine or bromine) may be added to the gas in the G-M tube; in the presence of such a quenching gas the positive ions do not cause the release of electrons when they strike the cathode. Most G-M tubes in use at the present time are of the self-quenching type; they contain argon, as the main filling gas, and a small proportion of a quenching material, e.g., ethyl alcohol.

Dead Time and Resolving Time

5.83. If properly quenched, either internally or externally, the voltage pulse from a G-M tube, as a function of time, has a shape similar to that shown at the left in Fig. 5.20. A second pulse arriving within the period from O to D will produce no effect; this period is called the *dead time* of the counter. It is caused by a positive space charge, due to the slow-moving positive ions, around the central wire anode. At successively later times, a second pulse of increasing voltage will appear, but only after the time R will it attain its full height. The time period DR is referred to as the *recovery time*. The resolving time of the G-M counter, which is related to the period OR, is generally longer than 2×10^{-4} sec, and so is some 200 or more times greater than the resolving time of a good proportional counter or ion chamber. The maximum rate of arrival of pulses which can be resolved by means of a G-M counter is thus about 5000 per sec.

5.84. Because the emission of nuclear radiations is invariably a random process, ionization pulses are not produced uniformly. There is always a possibility, therefore, even at relatively low pulse rates, that some pulses will arrive during the dead time of the counter. This means that the maximum practical counting rate, to be at all accurate, must be less than given above. In any event,

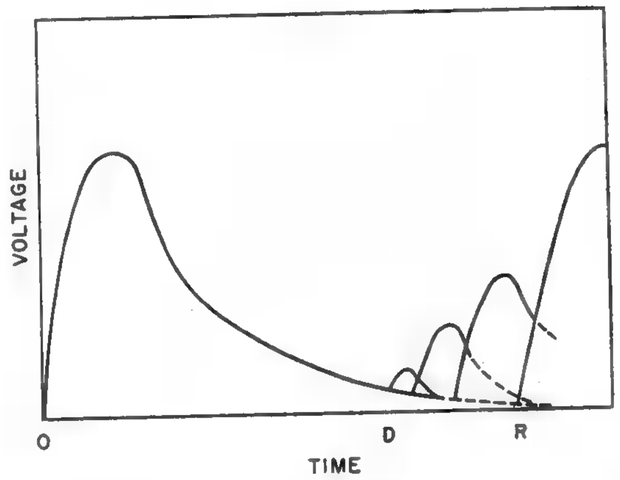


FIG. 5.20. Dead time and resolving time of G-M counter

it is necessary to apply a correction for "coincidence loss." This correction increases with the counting rate and with the resolving time (or dead time) of the tube.

SCINTILLATION COUNTERS*

IONIZATION IN SOLIDS AND LIQUIDS

5.85. All the instruments described above for the detection of nuclear radiations have been based on the production of ion-pairs in a gas, followed by their collection and recording in an appropriate manner. Nuclear radiations also produce ionization (and excitation) in their passage through solids and liquids, but the subsequent collection and measurement of the ion-pairs have proved difficult. The restriction to gaseous media has not been serious, except in connection with the measurement of gamma radiation. For this latter purpose an ionization medium of fairly high density and high atomic number is desirable if the detection efficiency is to be good, since the probability of interaction of the radiation with the medium is thereby increased. In the scintillation counter,

* For reviews see W. H. Jordan and P. R. Bell, *Nucleonics*, **5**, No. 4, 30 (1949); W. H. Jordan, *Ann. Rev. Nuc. Sci.*, **1**, 207 (1952); J. B. Birks, "Scintillation Counters," McGraw-Hill Book Co. Inc., 1954; S. C. Curran, "Luminescence and the Scintillation Counter," Academic Press, Inc., 1953.

which has developed rapidly during the past few years, a solid or liquid is used as the medium with which the radiation interacts.

5.86. It has long been known that certain solids and liquids, referred to as *phosphors*, emit light when exposed to nuclear radiations. In traversing the phosphor the ionizing particles or radiation cause the molecules of the material to become ionized or excited; these molecules then emit their excess energy in the form of light, each interacting particle giving rise to a flash, generally called a scintillation. This is the basic principle used in the scintillation counter.

DETECTION AND COUNTING OF SCINTILLATIONS

5.87. The great revival of interest in scintillation counters in recent years is due largely to two factors. First, there was the discovery that certain phosphors, e.g., organic compounds such as naphthalene, anthracene, diphenyl, etc., either in the solid state or in solution, or inorganic compounds, such as sodium iodide (activated with thallium), calcium and magnesium tungstates, etc., are transparent to the light they emit when exposed to nuclear radiations. As a result relatively large masses of the phosphor can be used; this insures a high probability of interaction with the radiation and a high detection efficiency. The second factor was the development of photomultiplier tubes whereby the effect of a scintillation can be greatly enhanced, thus permitting its observation and recording.

5.88. One method for combining the phosphor with a photomultiplier tube is shown in Fig. 5.21, the purpose of the aluminum foil being to act as a reflector of the light flashes. A particle of nuclear radiation or a gamma-ray photon entering the phosphor produces a flash of light, either directly or indirectly. This light falls upon the photocathode, on the inner surface of the glass envelope of the photomultiplier tube, where a number of photoelectrons are liberated. These are increased about a million-fold (or more) in passing through the tube, so that an appreciable voltage pulse is produced at the output stage. The pulse can be further amplified electronically and counted in the same manner, to be described below, as for other detecting devices.

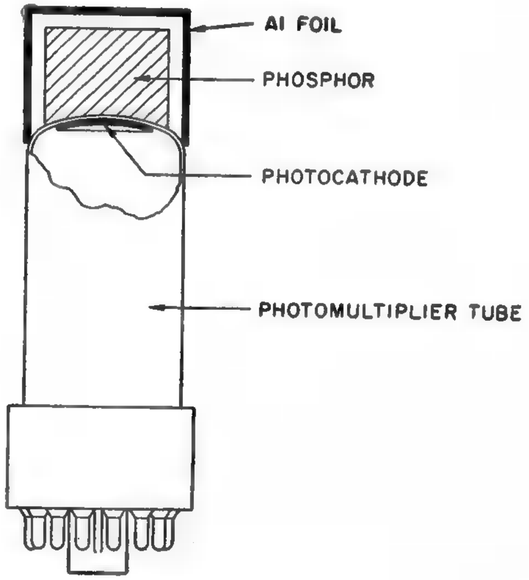


FIG. 5.21. Diagrammatic representation of scintillation counter

5.89. The chief advantages of the scintillation counter are its efficiency, resulting from the use of relatively large masses of phosphor, as already indi-

cated, and the high precision and counting rates which are possible. These latter attributes are a consequence of the extremely short duration of the light flashes, from about 10^{-9} to 10^{-6} sec, i.e., from 1 millimicrosec to 1 microsec. The intensity of the light flash and the amplitude of the output voltage pulse are proportional to the energy of the particle responsible for the flash. Consequently scintillation counters can be used to determine the energy, as well as the number, of the exciting particles (or photons).

5.90. In connection with reactor operations, scintillation counters have hitherto been mainly used for detecting and measuring low-level gamma radiation. Formerly, Geiger counters were largely employed for this purpose, but their efficiency is very low. On the other hand, scintillation counters are higher priced and require somewhat more complicated equipment than Geiger counters. By using phosphors containing lithium, scintillation counters can be adapted to neutron detection. Light flashes are produced by the helium and tritium nuclei resulting from the (n, α) reaction of the slow neutrons with lithium-6.

PULSE COUNTERS*

Binary and Decade Scalers

5.91. A mechanical recorder can register no more than about 10 pulses per sec, and this is much too slow for most purposes connected with the counting of ionizing particles. By means of an electronic *scaler*, however, only one pulse in 2, 4, 8 (or 2^n in general) or one pulse in 10, 100 (or 10^n in general) is permitted to pass on to the recorder; these are called "binary" and "decade" scalers, respectively.† The former are considered, on the whole, to be more reliable. In order to obtain the actual number of counts arriving in a given time period, the number recorded in that time is multiplied by the factor of the scaler. To this is added the number, less than this factor, still remaining in the circuit at the end of the counting period, as indicated by neon "interpolation" lights.

5.92. For reasons related to the statistics of counting (§ 5.97, *et seq.*), it is desirable, especially in connection with measurements which have to be compared with one another, always to count the same number of pulses. The time required is then inversely proportional to the intensity of the radiation. In some instruments the circuits are so arranged that an electric timer is started at the instant counting commences, and is stopped automatically after a prescribed number of pulses have been received.‡

* W. C. Elmore and M. L. Sands, *op. cit.*, Chapter 4.

† B. E. Phelps, *Electronics*, **18**, 110 (1945); W. A. Higinbotham, J. Gallagher, and M. L. Sands, *Rev. Sci. Inst.*, **18**, 706 (1947).

‡ See, for example, R. P. Ghelardi and C. H. Brown, *loc. cit.*

Counting-rate Meters

5.93. The type of counting described above is largely used, for laboratory measurements, when the time period required for the pulses to be counted is not of particular significance. If a rapid indication of the radiation intensity is required, as in reactor control, a *counting-rate meter* can be used in conjunction with a pulse chamber and its associated amplifier.*

5.94. The essential part of the circuit is a resistance-capacitance arrangement as shown in Fig. 5.22. The design of the charge generator is such that every pulse from the amplifier, exceeding a certain minimum voltage, trips a trigger circuit and thereby places a fixed charge, q, on to the capacitor C. This charge



Fig. 5.22. Block diagram of counting-rate meter circuit

would leak through the shunting resistance R, at a rate determined by the time constant (RC) of the circuit. The opposing effect of the charging of the capacitor by the arrival of pulses and the leakage through the resistance leads to an equilibrium state. If N is the average number of pulses arriving per second, the rate of gain of charge is qN, whereas the rate of loss is E/R, where E is the voltage produced across the capacitor (and resistance R). Hence, at equilibrium,

$$qN = \frac{E}{R} \tag{5.94.1}$$

so that

$$E = qNR. \tag{5.94.2}$$

Since q and R are constant, it is evident that the voltage E is proportional to N, the rate of arrival of pulses. The reading on the voltmeter is then a direct measure of the radiation field, e.g., the neutron density or the flux, in which the ionization chamber is placed.

5.95. Due to the random time of arrival of the pulses, the voltage E will fluctuate, the amount of fluctuation depending on the smoothing time constant RC and on the counting rate (§ 5.97). The root-mean-square fluctuation (ΔE) is given by

$$\frac{\Delta E}{E} = \frac{1}{\sqrt{2NRC}}.$$

The range of the instrument, i.e., the counting rate corresponding to full-scale reading of the voltmeter, is varied by changing R. Then as the counting rate,

* For descriptions, see A. G. Bousquet, *Nucleonics*, **4**, No. 2, 67 (1949); E. H. Cooke-Yarborough and E. W. Pulsford, *Proc. Inst. Elec. Eng. (Lond.)*, **98**, Part II, 191 (1951).

N, becomes smaller, for example, R can be increased proportionately so that $\Delta E/E$ remains constant. The amount of smoothing and speed of response are determined by the capacitance, C, of the capacitor.

5.96. For reactor control it is convenient to have a meter that indicates the counting rate on a logarithmic scale. This can be achieved by replacing the resistance R in Fig. 5.22 by a thermionic diode. As seen in § 5.53, within a certain current range, the voltage across such a diode is proportional to the logarithm of the current flowing through it. Therefore, from equations (5.56.1) and (5.94.1), recalling that $i = E/R$,

$$V = B \log kqN,$$

so that the reading on the voltmeter will be proportional to the logarithm of the pulse rate and, hence, of the neutron (or other radiation) density. An instrument of this kind is called a "log count-rate meter," abbreviated to "log CRM." If the voltage from the logarithmic circuit is applied to a differentiating system, the output can be used to indicate reactor periods, as in § 5.56.

STATISTICAL ERRORS OF COUNTERS *

Standard Deviation

5.97. All individual nuclear events appear to be random in character, although when a large number of such events are examined a definite probability pattern is evident. If any single radioactive nucleus were considered, it would be impossible to say when it is likely to emit a particle. But, for a system consisting of a very large number of nuclei of the same kind, the probability that any nucleus will expel a particle is determined by the decay constant of the given species (§ 2.5). Similar considerations apply to fission and other nuclear reactions. One of the consequences of the random nature of nuclear processes is that, if two or more identical counters are exposed to identically the same radiation source, the number of counts recorded on each will not necessarily be the same. If successive readings are taken on the same counter, they may differ somewhat.

5.98. According to probability theory as applied to purely random events, if $\bar{n}$ is the average of a large number of measurements, the probability P_n that n events will be recorded in any particular measurement is given by Poisson's law as

$$P_n = \frac{(\bar{n})^n e^{-\bar{n}}}{n!}. \tag{5.98.1}$$

The standard deviation, S,† i.e., the root-mean-square of the deviations of actual

* W. C. Elmore, *Nucleonics*, **6**, No. 1, 26 (1950).

† Fluctuations are sometimes expressed in terms of the *probable error;* this is the error for which there is an equal probability that it will or will not be exceeded. The probable error is 0.6745 times the standard deviation.

measurements from the average, is defined by

$$S = \left[\sum_0^\infty (\bar{n} - n)^2 P_n\right]^{\frac{1}{2}}. \tag{5.98.2}$$

Upon substituting the value of P_n given by equation (5.98.1), using Stirling's approximation for $n!$, and replacing the summation in equation (5.98.2) by integration, it is found that

$$S \approx \sqrt{n}.$$

The relative deviation S/n is then represented by

$$S/n \approx 1/\sqrt{n}. \tag{5.98.3}$$

5.99. If it is desired, for example, to determine a neutron flux (or other radiation) with an accuracy of 1 per cent, so that S/n is about 0.01, it follows from equation (5.98.3) that n should be 10^4. In other words, it would be necessary to count 10,000 ionizing events or pulses, if the result obtained is to be accurate within 1 per cent. The counting accuracy evidently depends on the total number of counts taken. This is an important point in favor of an instrument which records the time required for a specified number of pulses to be counted (§ 5.92).

Amplifier Noise and Background Radiation

5.100. A possible source of error in counting particles, as well as a limit to the sensitivity of a radiation detector, is due to what is called *amplifier noise;* it arises largely from the statistical nature of the electronic and ionic phenomena in the vacuum tubes upon which operation of the amplifier depends. Statistical fluctuations occurring in the early stages would be considerably amplified in later stages and might ultimately give rise to voltage pulses large enough to be counted. Amplifiers are consequently designed to give the maximum signal-to-noise ratio compatible with other requirements of the counter circuit.

5.101. Mention may also be made here of the *background radiation* as a source of extraneous counts, leading to errors in recording, although not statistical in origin. Even in the apparent absence of ionizing radiations, all counters of sufficient sensitivity will register the background radiation. This arises in part from cosmic rays* and in part from the presence of traces of radioactive matter in the air, in the construction materials of the counter, and in surrounding objects. Since these radiations cannot be eliminated completely, the background reading of the counter or rate meter is subtracted from all measurements.

* Cosmic rays owe their discovery to the observed continuous but slow leakage of charged electroscopes even when apparently shielded from ionizing radiation.

ACTIVATION METHOD FOR NEUTRON MEASUREMENT

Saturation Activity

5.102. An entirely different principle for the determination of neutron flux is widely used in preliminary reactor studies and also for standardization of neutron-sensitive ionization chambers and similar devices. It is also utilized, in a modified form, to indicate the need for safety action in some nuclear reactors (§ 5.126). This method depends on the fact, mentioned in preceding chapters, that certain elements are converted into radioactive species as a result of neutron capture. As a general rule, the reaction is of the radiative capture (n, γ) type, but this is not necessarily the case.

5.103. It was seen in § 2.202 that, when a neutron absorber is exposed to a constant neutron flux for a sufficient time to permit equilibrium to be attained, the equilibrium concentration of the resulting radioisotope is given by equation (2.202.1) as

$$\text{Radioactive nuclei per cm}^3 \text{ at equilibrium} = \frac{\Sigma\phi}{\lambda^*}. \tag{5.103.1}$$

In this expression Σ is the macroscopic *absorption* cross section of the neutron absorber, ϕ is the steady neutron flux, and λ^* is defined by

$$\lambda^* = \lambda + \sigma_p\phi,$$

where σ_p is the microscopic neutron absorption cross section and λ is the decay constant of the radioactive product. In all cases of interest for the present purpose, $\sigma_p\phi$ is negligible in comparison with λ. For example, σ_p may be 10^{-20} cm^2/nucleus and ϕ not more than 10^{14} neutrons/(cm^2)(sec), so that $\sigma_p\phi$ is 10^{-6} sec^{-1}, compared with useful values of about 10^{-2} to 10^{-3} sec^{-1} for λ; hence equation (5.103.1) may be written as

$$\text{Radioactive nuclei per cm}^3 \text{ at equilibrium} = \frac{\Sigma\phi}{\lambda}.$$

5.104. Suppose a substance having a volume V cm^3 is exposed in the manner described; the total number of radioactive nuclei at equilibrium is then $V\Sigma\phi/\lambda$. The rate of disintegration, in nuclei per second, is equal to the number of nuclei present, at any instant, multiplied by the decay constant (§ 2.5). It follows, therefore, that

$$\text{Rate of decay at equilibrium} = V\Sigma\phi \text{ nuclei/sec.}$$

If each decay is accompanied by the emission of one beta particle (or one gamma-ray photon), the equilibrium activity of the exposed specimen, as measured by the rate of emission of particles, is equal to the decay rate; thus,

$$\text{Activity at equilibrium} = V\Sigma\phi \text{ particles/sec.}$$

This equilibrium activity, expressed in terms of the rate of particle emission, is called the *saturation activity* (or *saturated activity*) of the given specimen. If the saturation activity is represented by A_∞, then

$$A_\infty = V\Sigma\phi, \tag{5.104.1}$$

so that it is seen to be proportional to the neutron flux.

Determination of Neutron Flux

5.105. In order to determine the neutron flux, e.g., in a reactor, a thin foil of neutron absorber is exposed for some time, so that it may be assumed that equilibrium is attained. A thin foil is used in order that the flux shall not be appreciably affected by the absorber and also in order that it may be nearly uniform throughout. The material must be one having a fairly high absorption cross section, which must be known, for the neutrons being observed so that A_∞ will be large enough to be easily determined. Further, the disintegration rate of the radioactive product should not be too large, so that it does not decay too rapidly while its activity is being measured. Materials which have been used for the purpose are indium, rhodium, silver, and manganese.

5.106. Upon removal of the activated foil from the neutron flux, the radioactive nuclei will continue to decay, but they are no longer being regenerated. If $A(t)$ particles/sec is the activity at any time t after removal from the neutron flux then, by equation (2.5.2),

$$A(t) = A_\infty e^{-\lambda t}$$

or, using equation (5.104.1),

$$A(t) = V\Sigma\phi e^{-\lambda t},$$

provided the saturation activity had been attained before the foil was taken out of the neutron flux. For practical purposes, Σ may be replaced by the value given by equation (2.113.2); the result is

$$A(t) = \frac{wN_a}{A}\sigma\phi e^{-\lambda t}, \tag{5.106.1}$$

where w, equal to $V\rho$, is the mass of the foil, A is the atomic weight of the foil element, and N_a is the Avogadro number. If the physical properties, A and σ of the foil material and λ of the radioactive product, are known, the neutron flux, ϕ, can then be calculated from the activity measured at time t after removal from the flux.

5.107. The activity of the foil after exposure is determined from the rate of particle emission as measured by a suitable counter of the ionization or scintillation type. It is important in this connection that what is called the *geometry* of the counter should be known. It is a measure of the proportion of the particles emitted by the activated foil which lead to countable events. The total rate of particle emission can then be estimated, and from this the neutron flux can be calculated by means of equation (5.106.1).

5.108. If, for various reasons, it is suspected that the foil has not been exposed long enough for equilibrium to be attained, so that the activity is less than the saturation value for the given flux, use may be made of equation (2.203.2). In the present case this becomes

$$A(t) = A_\infty(1 - e^{-\lambda T})e^{-\lambda t}$$
$$= \frac{wN_a}{A}\sigma\phi(1 - e^{-\lambda T})e^{-\lambda t},$$

where T is the time of exposure to the neutrons and t is the time after removal from the neutron flux at which the activity of the foil is measured.

5.109. It is important to note that the neutron flux determined in the foregoing manner is not necessarily the total flux, but that of the neutrons to which the value of σ applies. The method is thus particularly useful for thermal neutrons, where it is possible to ascribe a more or less definite average absorption cross section (§ 2.129).

INSTRUMENTS FOR REACTOR CONTROL*

Neutron Flux Determination

5.110. The neutron flux at any point is proportional to the fission rate and hence also to the power level of the reactor in the region where the flux is measured. Because instruments can be made to respond to changes in the flux very rapidly, they provide a virtually immediate indication of local power variations. For control purposes, therefore, they have, in certain respects, advantages over more conventional devices for indicating power levels. During the early stages in the start-up of a reactor, the power output is so small that instruments of the latter type would not register. Under those conditions recourse must be had to ionization chambers and similar neutron detectors, in any event.

5.111. The safe operation of a reactor requires that the neutron flux (or fission rate) be known over a very large range; for example, the flux may vary by a factor of 10^{10} or more between start-up and operating level. During start-up it is quite possible for the reactor to become critical and for the period (§ 4.20) to be very short while the neutron flux is still low. If this occurred, the reactor power might increase so rapidly that dangerous levels would be reached before steps could be taken to reduce the fission rate. Short periods should be avoided for safety reasons, and so the fission rate must be measured from the initial level at start-up to the operating level, if possible.

5.112. No single instrument, nor even any one type of instrument, can be used for satisfactory measurement of neutron flux in a reactor over the whole possible range; as a result, it has been found convenient to divide this range into four regions. Although these regions are not exactly defined, and will vary to some extent from one reactor to another, they are approximately as indicated in Fig. 5.23. The right end of the scale, indicated by unity, represents full power of the reactor and the various negative powers of ten are the respective fractions of full power. In the *power range*, the power (or flux) is from full power to 10^{-2} of full power; the *period range* covers from 10^{-2} to 10^{-6} of full power; the *counter range* is from 10^{-6} to 10^{-11}; while the *source range* is that below 10^{-11} of full power. No suitable method is available for indicating the neutron flux in the source range, but none is needed because a source is usually present which brings the flux up into the counter range.

* D. Cochran and C. A. Hansen, Jr., *Nucleonics*, **5**, No. 2, 4 (1949); J. D. Trimmer and W. H. Jordan, *ibid.*, **9**, No. 4, 60 (1951).

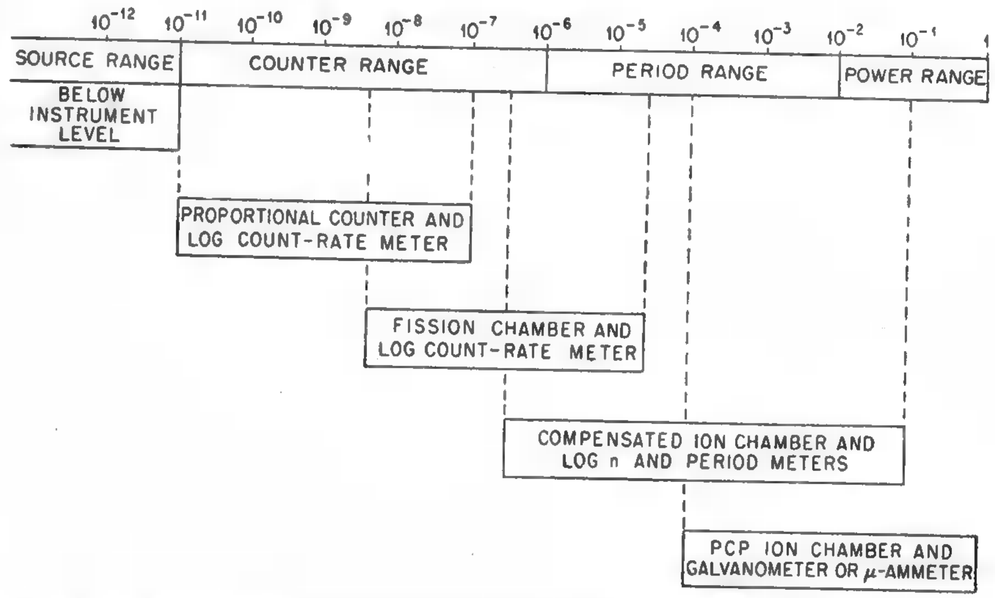


Fig. 5.23. Reactor start-up and operating ranges with appropriate instrumentation

5.113. The ideal instrument for reactor control should be capable of detecting neutrons in the presence of strong gamma radiation; in addition, it should be accurate and respond rapidly to changes in neutron flux. These requirements are sometimes in conflict. When the neutron flux is low but the gamma radiation high, an ionization chamber or proportional counter in conjunction with a pulse-rate counting system is desirable because it is capable of discriminating between the large pulses due to neutrons and the small pulses produced by gamma rays. However, since the neutron flux is small, the rate of arrival of pulses is small and a long integrating time is necessary if accurate readings are to be obtained (cf. § 5.99); the response will then be slow. If the instrument is designed to respond rapidly, the readings will be inaccurate. At higher neutron fluxes the response of a counting-rate meter would be more rapid, for a given accuracy, but a limit is set by the resolving time of the system. Fortunately, for large rates of arrival of pulses a current-measuring (or integrating) type of

instrument is capable of accuracy and rapid response. The effect of gamma radiation can then be overcome by the use of a compensated chamber.

5.114. During the early stages of the restart of a reactor after shut-down, the gamma radiation field is high while the neutron flux is relatively low. A pulse-type ionization chamber (or proportional counter), with appropriate bias on the counter, is the best device for measuring the neutron flux under these conditions. A fission chamber is better in this connection than one containing boron, because the ionization pulses are much larger in the former case, thus facilitating discrimination against gamma radiation. Since the neutron flux is small, the number of ionizing events in the chamber will be small and the accuracy of the reading will be low. Fortunately, this does not matter in the circumstances, and so, at the commencement of the start-up of a reactor, approximate values of the neutron flux are usually indicated by means of a pulse-type fission chamber or a proportional counter with boron trifluoride, combined with a counting-rate meter. It is for this reason that the description "counter range" has been applied to the region in which the flux ranges from about 10^{-11} to 10^{-6} of full power.

5.115. Since the neutron flux during start-up may vary over several powers of ten, in the counter range, it is convenient to use a meter that indicates the counting rate due to the neutrons on a logarithmic scale. Such an instrument is the log count-rate meter (§ 5.96); meters reading from 1 count per sec to 10,000 per sec, i.e., from zero to four on the logarithmic scale, are used with many reactors. Even greater ranges of neutron flux can be measured by moving the fission chamber or detector into a part of the reactor where the flux is lower. The withdrawal of the chamber from the high-flux region is an advantage, in any event, since the problems of radiation damage and induced radioactivity of the chamber materials are thereby avoided.

5.116. Because of the resolving time of the pulse chamber, the maximum rate of arrival of pulses, for which satisfactory readings are obtainable, is about 10^4 or 10^5 per sec. When this condition is reached, the neutron flux is large enough to make possible the use of an integrating ionization chamber, thus taking advantage of its more rapid response. As seen earlier, satisfactory operation of such a chamber commences when the ion current is about 10^{-10} amp, corresponding to about 10^4 particles (or pulses)/sec, and this is approaching the point where the pulse chamber ceases to be effective. Since the number of ionizing events in the chamber is still relatively small, the accuracy and steadiness of the current will not at first be great, but both increase with increasing neutron flux.

5.117. In the period range the main concern is the rate at which the neutron flux is increasing, and so the reactor period is determined by means of a meter which indicates the time-derivative of the logarithm of the neutron flux (§ 5.56). The magnitude of the flux is recorded simultaneously by means of a log n meter (§ 5.52). Since it is desirable to know the reactor period at as low a power (or neutron) level as possible, so that dangerously short periods may be detected, a

period meter is used in conjunction with a compensated ionization chamber, such as the one described in § 5.39. In this way the effect of the large gamma-ray intensity is eliminated.

5.118. In the power range, i.e., from about 10^{-2} of full power to full power, accuracy and sensitivity are the essential requirements of the indicating device, especially if automatic control is employed. For this purpose an ionization chamber is used, but, instead of logarithmic amplification, the ion current, after necessary linear amplification, is recorded directly on a galvanometer or microammeter (see § 5.43) appropriately calibrated to read neutron flux or reactor power. The output of such an instrument may be matched against a prescribed value, representing the desired full power of the reactor, and the difference applied to the control-rod actuating mechanism. As long as any difference exists, the neutron flux (and power level) is permitted to increase at a controlled rate, but when full power is reached the increase is halted automatically (see Chapter VI). The reactor is then maintained at the desired power level in an analogous manner; transient changes in flux, as indicated by means of the ion chamber, are compensated for by prompt, automatic movement of a regulating rod.

5.119. Because of its very rapid response, the parallel-plate (PCP) ionization chamber, referred to in § 5.36, is often used for automatic control. It is true that the ion current includes some contribution from gamma radiation, but this is not important since it can be allowed for in the reference voltage (or current) against which the output of the chamber is matched. For the same reason as given above, the parallel-plate chamber is recommended for the actuation of safety circuits.

5.120. In the foregoing discussion it has been assumed, for convenience, that the counter, period, and power ranges are more or less distinct. This is, of course, not the case, and the various instruments used overlap the boundaries, as indicated in Fig. 5.23, thus providing continuity of indication of neutron flux or power level. The log CRM instrument, for example, has a range of 10^4, part of which is covered by the log n meter with a range of about 10^6; finally, the microammeter or galvanometer used in the power region is usually provided with shunts which give an operating range of 10^4 or 10^5.

Instruments for Special Purposes

5.121. The instrumentation problems considered above have referred in particular to devices using ionization produced indirectly by neutrons to indicate the neutron flux (or power level) of the reactor. In addition, a number of devices of a somewhat novel character, based on different principles, have been developed for various purposes. One of these is the neutron thermopile,* which is a sturdy and reliable instrument not much larger than a fountain pen. It contains some forty thermocouples, alternate junctions being coated with

* J. Barbaras, *et al.*, Report AECD-2075; A. C. Lapsley, *Nucleonics*, **11**, No. 5, 62 (1953).

boron. The heat generated when the boron interacts with neutrons is capable of producing an appreciable voltage. Thus, in a thermal flux of 10^{12} neutrons/(cm^2)(sec), the output is approximately 10 millivolts. Since thermocouples have a low impedance, it is not necessary to maintain high insulation resistances, such as are required for ionization chambers. Their chief disadvantage is a relatively long response time, of the order of seconds, so that they are not recommended for control purposes. Nevertheless, neutron thermopiles can play a part in reactor instrumentation.

5.122. A single neutron detector placed at a particular point in a reactor indicates only the flux (or power level) in its immediate vicinity, but does not give a good idea of the average power level of the reactor as a whole. The flux in the region of the detector may be considerably influenced by absorbers in the vicinity and by the position of the rods used to control the reactor. A much more satisfactory indication of the over-all power level would be provided by indication of the neutron flux averaged over the reactor volume. This can be approximated by placing a number of detectors at various points in the reactor and connecting their output terminals in an appropriate manner. The relatively large volume occupied by an ionization chamber makes its use as a detector impractical in this connection, but boron thermopiles are small enough to be very suitable for the purpose of recording the average reactor power level.

5.123. Boron thermopiles can be used as a part of the safety system of a reactor even though their response is slow. Certain corrective actions of the control rods can be initiated automatically when the average power level, as indicated by the thermopile, exceeds the permitted full operating level of the reactor by a specified amount (see Table 6.4). In addition, boron thermopiles have been employed for monitoring neutron flux at a given point in the reactor, for example, where an experiment is in progress. Knowing that the reactor power has been held constant is not sufficient for certain experiments, because of the local "shading" effects, etc.

5.124. The induced activity produced by neutrons in certain materials can be used to determine neutron flux (§ 5.102), but the method has not found much application in reactor control for two reasons. First, the information is generally not continuous, and, second, it is not available promptly. There is a possibility of using the material to be activated in the form of an endless belt or wire which can be circulated continuously through the reactor. The idea has not been applied because of the large time lags, but it has the merit of complete discrimination against gamma radiation.

5.125. A scheme involving essentially the same principle, however, has been employed in reactors using water as coolant. The water flowing through the reactor becomes activated as a result of the (n, p) reaction of the fast neutrons with the O^{16} in the water (§ 2.96). The product, N^{16}, decays with a half life of 7 sec, emitting a high-energy ($\sim$6 Mev) gamma ray in the process. An ionization chamber is then used to detect the gamma radiation of the effluent water.

Since the water is thoroughly mixed before reaching the detector, the measured activity is a good indication of the average (fast) neutron flux and of the over-all power level of the reactor. The method cannot be employed over a wide range of neutron flux, because of other activities of long life built up in impurities in the water, but it can be used, like the boron thermopiles, to initiate corrective safety action.

5.126. Although the various devices already described, e.g., fission chambers, proportional counters, compensated ion chambers, boron thermopiles, etc., can be calibrated to indicate power level, such calibration must depend on direct determination of reactor power from measurements of rate of flow of coolant and of the rise in temperature. More or less conventional instruments can be used for the indication of both these quantities, and the power output is then given by their product.

5.127. Instead of actually performing the multiplication of coolant flow and temperature rise, it is a simple matter to design a device for indicating this product directly in watts (or kilowatts). In the bridge circuit represented in

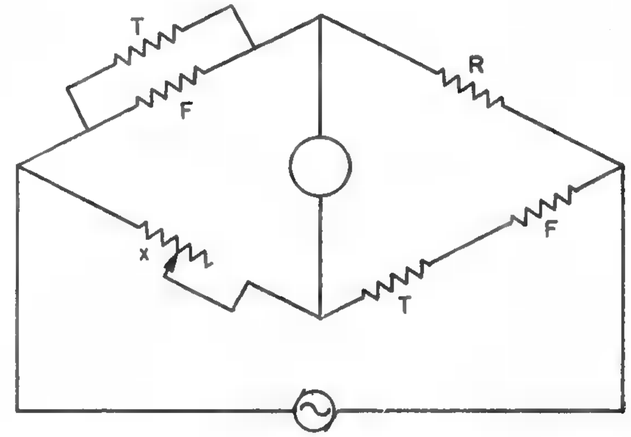


FIG. 5.24. Bridge circuit for direct determination of reactor power

Fig. 5.24, the resistances T and F are proportional, respectively, to the temperature rise and to the flow rate of the coolant. If R is a fixed resistor, and the bridge is kept balanced automatically by adjustment of the resistance x, it can be readily shown that the balance condition is

$$x = \frac{TF}{R}.$$

The value of the resistance x is then a direct measure of the power level, since the latter is proportional to the product TF. A meter based on this principle should not be used in the automatic control system, because of its slow response to changes in the fission rate, but it can be included in safety circuits.

Symbols Used in Chapter V

A	atomic weight
A	activity (disintegrations (or particles)/sec)
$A(t)$	activity at time t after removal from neutron flux
A_∞	saturation activity (for given flux)
B	constant
C	capacitance
E	voltage (across capacitor)
e	base of natural logarithms
F	flow rate
i	current (also at time t)
i_0	current at time $t = 0$
i_r	current reading (on meter)
k	constant
N	number of pulses/sec (or counting rate)
N_a	Avogadro number
n	neutron density
n	number of events
$\bar{n}$	average of large number of measurements
P_n	probability of n events being recorded
q	charge on capacitor
R	resistance
S	standard deviation
T	temperature rise
T	reactor period (stable)
T	time of exposure to neutrons
t	time
t	time after removal from neutron flux
V	volume of material exposed to neutrons
V	voltmeter reading
V	voltage (also at time t)
V_0	voltage at time $t = 0$
w	weight of foil
λ	radioactive decay constant
λ^*	effective decay constant ($\lambda + \sigma\phi$)
ρ	density
Σ	macroscopic (capture) cross section
σ	capture cross section of exposed material
σ_p	capture cross section of (n, γ) product
ϕ	neutron flux

PROBLEMS

1. It is desired to use a parallel circular-plate (boron-10 coated) ionization chamber to control the operating power of a graphite-reflected, cubic reactor having an average thermal flux of 10^{13} neutrons/(cm^2)(sec). Roughly at what distance from the reactor core face might the instrument be located?

2. An indium foil, weighing 0.050 gram, was exposed to thermal neutrons for exactly 1 hr; the beta activity 2 hr later was found to be 2.42×10^5 particles emitted per sec. Determine the value of the neutron flux. (The atomic weight of indium may be taken as 115 and the capture cross section for thermal neutrons as 190 barns; the half life of indium-116, the (n, γ) product, is 54 min.)

3. What would have been the observed activity 3 hr after removal from the neutron flux if the foil had been exposed until the saturation activity had been virtually attained? About what exposure time would be necessary to achieve this, allowing for a 1 per cent deviation?

4. Assuming a 5 per cent counting efficiency, due to the "geometry" of the counting system, about how long should the counting in Problem 2 be continued if an accuracy of 1 per cent is desired? Is there an appreciable decrease in the activity (rate of particle emission) during this time?

5. A nuclear reactor power plant uses water at high pressure and temperature as coolant; after leaving the reactor the water is circulated through an external heat exchanger where steam is produced. Make as complete a list as possible of the instruments that might be used in the plant and state their purpose in each case.

Chapter VI *

CONTROL OF NUCLEAR REACTORS

GENERAL FEATURES OF REACTOR CONTROL †

Basic Principles of Control

6.1. Like other devices for the conversion of energy, such as a steam boiler or an internal combustion engine, a nuclear reactor must have a suitable control system, the purpose of which is to provide smooth and steady operation under the circumstances for which the device was designed. Methods must be available for starting the reactor, for bringing the power output to the desired level, to maintain it at that level, and then to shut down the reactor when necessary. In addition, as with other energy converters, safety devices must be provided to prevent damage in case of an accident. The control (or operation) of a nuclear reactor is thus the same, in principle, as the operation of other means for making energy available from a fuel, such as coal or oil. The situation may be summarized by stating that the subject of reactor control concerns the devices and methods needed to permit operation of the reactor and to provide adequate safeguards in the event of an emergency.

6.2. An important difference between the control of a reactor and the control of other energy converters arises from the nature of the fuel. With but few exceptions, in the nuclear reactors which have so far been constructed or designed, the fuel supply, namely, the fissionable material, cannot be continuously replaced as it is consumed. It is consequently necessary that such a reactor have included in its construction all the fuel that is likely to be required for the production of a predetermined quantity of energy. In the case of mobile reactors, such as for use on shipboard or for propulsion of an aircraft, this may be quite considerable, since sufficient fuel should be included to permit continuous operation over extended periods.

* Material contributed by S. Glasstone, W. H. Jordan, and J. D. Trimmer. Reviewed by T. E. Cole.

† For general reviews, see L. A. Ohlinger, *Nucleonics*, **6**, No. 1, 10 (1950); J. M. Harrer, *ibid.*, **6**, No. 3, 58 (1950); J. D. Trimmer, and W. H. Jordan, *ibid.*, **9**, No. 4, 60 (1951); M. A. Schultz, *Elec. Eng.*, **72**, 505 (1953); R. V. Moore, *Proc. Inst. Elec. Eng.* (*Lond.*), **100**, Part I, 90 (1953).

6.3. A nuclear reactor cannot be used to release energy continuously unless it always exceeds the critical size for the particular fuel, shape, etc. For this reason a power reactor must include excess fuel, and so it must obviously have latent reactivity to an appreciable extent at the time it commences operation. In addition, other factors, such as the need to override the effect of fission product poisons and of temperature (see Chapter IV), require that additional reactivity be built into the reactor. Because of the high reactivity, the neutron flux (or density) and, hence, the fission rate and the power are capable of an extremely rapid, and dangerous, increase in a very short time. An essential aspect of reactor control thus lies in the precautions to be taken to prevent an excessive rate of increase in neutron flux when the power level is being raised.

6.4. Since the power level of a reactor is virtually proportional to the neutron flux, the obvious basis for reactor control is to vary the effective multiplication or reactivity (§ 4.17). If the effective multiplication factor is greater than unity, the reactor is supercritical, and the power level will increase continuously. Upon decreasing the factor to unity, so that the reactor is just critical, the power output will remain constant (apart from transient changes) at the level attained at that time. Finally, by making the reactor subcritical, that is, by reducing the effective multiplication factor below unity, the power level will be decreased.

Methods of Control

6.5. There are four general methods which can be used to vary the effective multiplication factor (or reactivity); they involve addition or removal of (1) fuel, (2) moderator, (3) reflector, or (4) a neutron absorber. Each one of these methods, or a combination of them, has been used or proposed for the control of nuclear reactors already in existence or under construction.

6.6. The control procedure which has hitherto been widely employed, because of its simplicity and convenience, is the insertion or withdrawal of a material, such as boron or cadmium, which has a large capture cross section for thermal neutrons. Rods or strips of boron steel (or boron carbide) or cadmium in some form constitute a very effective means of reactor control. It was used exclusively in the original Chicago "pile" (CP-1) and in its reconstruction (CP-2), and is employed in the Oak Ridge (X-10) reactor and others. The control rods may be within the core of the reactor, as in the instances just mentioned, or they may be located in the reflector, as in the earliest (LOPO) form of the Los Alamos homogeneous reactor (Water Boiler).

6.7. The chief disadvantage of control by a strong absorber is the resulting loss of neutrons. This is not serious in systems having small excess reactivity and which have a low power output, for during operation the control rods are almost entirely withdrawn. However, such would not be the case for reactors having considerable built-in reactivity to take care of fuel depletion, fission product poisoning, etc., and which have high neutron fluxes. An improvement in neutron economy can be achieved by combining motion of the core material,

i.e., fuel and moderator, with that of an absorber. The lower portion of a coarse-control rod may be constructed of the same material as the reactor core, whereas the upper portion contains cadmium. When such a rod is lowered, so that the neutron absorber is inserted, some of the core is removed at the same time, thereby bringing about a further decrease of reactivity.

6.8. Instead of using a wasteful absorber, such as cadmium or boron, there is a possibility that the control rods could be made of an absorber, such as uranium-238 (or natural uranium) or thorium, which would produce a fissionable material — plutonium-239 or uranium-233, respectively — as the result of neutron capture and subsequent beta decay (§ 1.59, *et seq.*). Alternatively, the absorption of neutrons by the control rod could be used for the production of certain desired isotopes. It is to be expected that, ultimately, control by absorption in non-productive nuclei will be eliminated.

6.9. In a fast-neutron reactor control by means of a neutron absorber is not generally satisfactory because of the low capture cross sections for neutrons of high energy. The reactivity can, however, be changed by removal of fuel material from (or by addition to) the core or by movement of the reflector. In the Los Alamos fast reactor ("Clementine"), now dismantled, control was achieved by means of rods of natural uranium metal which were normally part of the reflector. It should be noted that, when fuel or reflector is used for control purposes, its direction of motion will be opposite to that of an absorber. In order to decrease the reactivity, fuel or reflector must be removed, whereas an absorber, on the other hand, must be inserted. This suggests the possibility of control by combining insertion of an absorber with removal of part of the reflector in a manner similar to that described for the core in § 6.7.

6.10. Another method of control is available when the moderator is a liquid. In the Canadian (NRX) high-flux reactor, for example, a certain degree of coarse control is achieved by altering the level of the heavy water used as moderator.

Source Control

6.11. Although it is at present of relatively minor importance in reactor control — except in certain experimental work and other cases — brief mention will be made, for the sake of completeness, of the possibility of reactor control by variations in source strength. In a subcritical reactor, where the neutron level is not very much greater than that provided by the source, control by changing the latter is feasible, provided the subcritical multiplication is not large. To obtain large multiplication, however, the reactor would have to be so near criticality that a reactivity control, e.g., a control rod, would also be needed. To control by a source alone, it would be necessary either to have a very strong source or else to operate at a relatively low power level. Source control should be quite good in speed, smoothness, and fineness. A modulated-strength, fixed-position source inside a reactor would also be ideal from the standpoint of neutron economy.

The Operator, Automatic and Load Loops

6.12. In order to understand some of the over-all features of reactor control, it is of interest to consider a simple form of nuclear reactor with extremely small reactivity, such as CP-1 or CP-2, operating at essentially zero power, and thus requiring no coolant. Provided an instrument is available to indicate the neutron flux, such a reactor could be controlled entirely by manual operation. In general, however, an appreciable expenditure of power is required to move the control rods, and the human operator can be aided in this respect by suitable motors.

6.13. Although operator control is feasible in certain instances, it is not always possible; neither is it advisable to rely entirely on human operation. Emergency safety action, for example, should be made automatic. Thus, if an instrument indicates that the neutron flux is rising at a dangerously rapid rate, or if the power level is appreciably higher than that for which the reactor was designed, an operator may not be able to act quickly enough to decrease the reactivity in time to avoid an accident. In circumstances such as these, control rods should be moved automatically to rectify the situation. Ideally, too, it would be advantageous if automatic devices were available for raising the power output of the reactor to the required level and for maintaining it there. As will be seen later, this aim has been achieved to a considerable extent.

6.14. A block diagram depicting some of the general aspects of reactor control is given in Fig. 6.1. Arrowheads indicate the direction of flow of information, signal, or effect; broken lines imply flow of information, while solid lines represent power. Following the lines representing the "operator loop," it is seen that the reactor affects certain instruments which pass information to the operator. The latter receives the information and, in turn, exerts appropriate action on, or conveys a signal to, the control block. It is then the function of the control

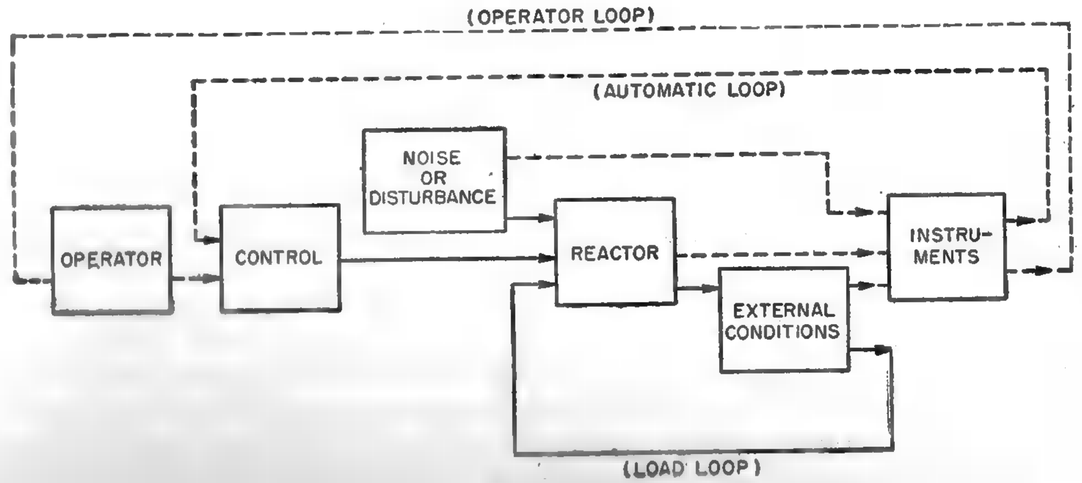


Fig. 6.1. General block diagram of reactor control system

block to exert the desired control action on the reactor, usually with the expenditure of power from an external source. In the "automatic loop" the information received from the reactor by the instruments is fed directly into the control block, the operator being by-passed.

6.15. A third loop, shown in Fig. 6.1 as the "load loop," is intended to represent the interaction of the nuclear aspects of the reactor, i.e., the fuel, moderator, and reflector, with the external conditions, or non-nuclear aspects. Among the latter may be included the status of equipment, such as a power plant, designed to be closely associated with the reactor, and of process facilities, such as water, air, and electricity, that may be essential to its operation. The external conditions may be regarded as constituting, in a general sense, the "load" on the reactor. The closed cycle in the block diagram implies that the load can affect the reactor, which in turn influences the load. The changes in the external conditions are indicated on suitable instruments, and information is fed back to the control block via operator and automatic loops, so that any necessary action may be taken.

6.16. An important aspect of the load loop is found in the interaction of the reactor and its cooling system. The heat released inside a reactor goes partly into heating the reactor itself and partly into the thermal load. In an equilibrium condition, all the heat goes to the load. Depending on the power level and, especially, on the power density of the reactor, the thermal load is a more or less elaborate cooling system designed to permit rapid transfer of heat from reactor to coolant. The coolant flow may be open-ended, as the flow of a gas into the atmosphere, e.g., in the X-10 and Brookhaven reactors, or of water into a river, e.g., the Hanford reactors; or it may be a closed circulating flow of liquid, e.g., when heavy water, liquid metal, or pressurized water is the coolant, or of a gas, e.g., nitrogen under pressure in the French reactor at Saclay. In any case, the heat exchange between coolant and reactor is an element of the control problem, as was indicated earlier.

6.17. In addition to the three closed loops described above, it will be observed that Fig. 6.1 includes a block labeled "noise or disturbance." These two terms as used here are essentially synonymous; the only reason for giving both is because of their more limited customary implications. "Noise" has been widely used beyond its acoustical meaning to signify relatively fine-grained, high-frequency, random fluctuations in any physical quantity. The term "disturbance," on the other hand, usually implies relatively gross, unexpected, or undesired effects. The label "noise or disturbance" is thus intended to include all undesirable effects on the reactor. As indicated on the block diagram, one method of counteracting some kinds of noise or disturbance is to use instruments to detect them as soon as possible and to initiate corrective control action.

6.18. Although the operator, automatic, and load loops have been shown as closed, they are actually open to some extent. For example, the operator brings into the control pattern a factor not shown on the block diagram: this is his concept of how the reactor should be operated. Depending on how the total responsibility is divided between the automatic loop and the operator loop, the operator is more or less free to put his concept into practice. In any case, the operator's concept, together with the concepts built into the automatic loop by the designer, determine the over-all performance of the total control system. It is clear, therefore, that the system is not altogether closed or isolated, since information, in the form of instructions or criteria of performance, enters from outside.

6.19. Information also leaves the system in various ways. For example, the instruments may furnish information about the reactor or the load to persons or places other than the operator or the control block. This aspect of the subject, however, has no immediate bearing on the control of a reactor and so it will not be considered here. In the same category is the information obtained from health physics instruments used for the monitoring of maintenance and other operations connected with the reactor.

6.20. The reactor system is also open with regard to energy; this leaves the system mainly via the load. In several isotope-producing and experimental reactors most of the energy is carried off by the coolant, such as air or water, which flows away. In a power-generating reactor, a part of the energy is delivered to the useful load. There is also energy input, mainly in the form of the fuel going into the reactor itself. In addition, as implied by the solid line representing the output of the control block in Fig. 6.1, an external source of power is required to amplify the weak signals from operator or instruments so as to cause effective action in the reactor. There are similar, though less important, energy inputs to other blocks in the diagram, e.g., to the instruments.

6.21. The discussion of the three loops — operator, automatic, and load — and their interaction constitutes an essential topic in reactor control. To some extent the treatment can be in mathematical terms; to a large degree, however, it must be based on experience, either with actual reactors or with reactor analogues or simulators (§ 4.56, *et seq.*). In reviewing the individual blocks in Fig. 6.1, it will be noted that the various aspects of the reactor and of instruments have been examined in earlier portions of this book. Although further reference to these matters will be made, the discussion in this chapter will center about the control block, mainly in terms of the operator loop, the automatic loop, and the interactions between them.

DESIGN OF THE CONTROL SYSTEM

Specification of the Control System

6.22. An important factor — perhaps the most important factor — underlying the whole treatment of control is the purpose and design of the reactor to be controlled. Some typical purposes for which reactors are designed are as follows: experimental and teaching purposes, isotope and plutonium production, genera-

tion of electric power in a stationary plant, aircraft power plant, or submarine power plant, and breeding of fissionable material. A given reactor may, of course, serve more than one purpose, but no matter what its function, this must motivate control design just as it motivates other features of reactor design. The controls for any reactor system cannot be properly designed until the required purposes and, therefore, the criteria of performance of the system are clearly understood.

6.23. Although the details of control design will vary from one type of reactor to another, there are nevertheless certain general requirements which all control systems should meet. In any given reactor the control action should have sufficient range to cover all contingencies and sufficient delicacy of action to permit fine adjustment. It should respond quickly to activating signals, without itself creating undue disturbance. The control system should be simple and foolproof in action, thus assuring a maximum of reliability. It should be designed so that there will be adequate safeguards in the event that any part should fail.* Ideally, too, it should be as economical as possible of neutrons.

6.24. Specification of a complete control system can thus be discussed in terms of three main requirements: range, accuracy, and efficiency. *Range* is concerned with the total amount of reactivity which must be at the command of the system; this obviously affects both operation and safety. *Accuracy* of operation control refers to the closeness with which the actual behavior of the reactor responds to the wishes of the operator or to the formula of the control system. In safety action, the term "accuracy" may be used to imply the promptness with which safety action follows upon the need for such action. Reliability may be considered as an aspect of accuracy. *Efficiency* is an over-all, combined measure of neutron economy, power economy, and economy of operation. Qualitatively, it may be said that an efficient control system works unobtrusively, using this word in its widest sense.

Types of Control Rods†

6.25. Before considering these specifications in further detail, it may be noted that to achieve both range and accuracy might prove difficult if a single type of control rod were used. Consequently most reactors have rods with different functions or serving different purposes. There are, first, the so-called *shim rods* for coarse control. They are used to bring the reactor approximately to the desired power level when the system is started up. The shim rods must, therefore, have a fairly large reactivity equivalent, the actual amount depending on the particular reactor as will be seen later. Because the rate of increase of reactivity must be closely regulated, the driving mechanism must be such that the shim rods cannot move at high speed during start-up.

* The dictum that component parts should be designed to "fail safe" applies not merely to controls, but to all aspects of reactor design, in so far as possible.

† The term "control rod," as employed here and subsequently, applies quite generally to either fuel, moderator, reflector, or absorber, the movement of which is accompanied by a change in the reactivity of the reactor.

6.26. When the desired power level of the reactor is approached, the shim rods are returned to such a position as to make the reactivity very small, that is to say, the effective multiplication factor is reduced almost to unity. The task of bringing the reactor up to the operating level and of maintaining the power essentially constant by counteracting the effects of rapid, transient changes, then passes to the *regulating rods*. The reactivity equivalent of these rods may be quite small, but in general they should be capable of moving rapidly so as to provide quick response to changes affecting the reactor. By keeping the reactivity equivalent small and, in some reactors, by limiting the distance over which they can move, the regulating rods are unable to produce any dangerous increase of reactivity.

6.27. As a general rule, the reactivity equivalent of a regulating rod should not exceed the fraction of delayed neutrons, e.g., 0.0075 (or 0.75 per cent) when uranium-235 is the fissionable material. Then, if the reactor is critical, complete withdrawal of the regulating rod, due to operator error or to failure of the automatic control system, would not permit the system to become prompt critical (§ 4.46).

6.28. If a high-flux reactor operates continuously at constant power, a gradual withdrawal of the regulating rod is necessary to compensate for the decrease in reactivity due to fuel depletion, accumulation of poisons, etc. When the rod reaches the end of its range, it must be reinserted into the reactor while a shim rod is withdrawn to an equivalent extent. Therefore in some reactors the shim rods and regulating rods are interlocked. An alternative to this procedure is to make use of the idea of shim control by suitable distribution of a "burnable" poison throughout the reactor core. If the consumption of the poison, due to neutron capture, occurs at a rate which largely compensates for the fuel depletion, the gradual withdrawal of the regulating (or shim) rods can be almost entirely avoided.

6.29. Finally, there are the *safety rods*, the purpose of which is to shut down the reactor quickly in the event of an emergency; this is generally referred to as "scram." Safety rods must, obviously, be capable of moving very rapidly, and their reactivity equivalent must be appreciably greater than the maximum excess of reactivity built into the reactor. In some reactors the same rods are used as shim rods and safety rods. While both types have high reactivity equivalents, it has been seen that the former must move slowly while the latter must move rapidly. These apparently contradictory requirements can be met by attaching the rods to the drive mechanism by means of magnetic clutches. When the clutches are energized the rods act as shim rods and can move only at a predetermined slow rate. In the event of an emergency, however, the clutches are de-energized and the rods fall rapidly, under the influence of gravity

(and sometimes of the pressure of the cooling water), to fulfill their safety function.

6.30. In addition to the control rods described above, most reactors, especially those of large dimensions, have what is called a "back-up" safety device. This is intended for use in an extreme emergency, such as an earthquake, when distortion of the reactor system would prevent normal motion of the safety rods. In the Oak Ridge X-10 reactor, the back-up safety consists of boron-steel shot which, in the event of an emergency, can be run into holes in the reactor. The Brookhaven reactor has a liquid neutron absorber, trichlorobenzene, in addition to shot, for the same purpose. Homogeneous reactors using a liquid medium, e.g., one form of the Water Boiler, can be fitted with "dump" valves so that fuel solution can be rapidly removed from the core into vessels having noncritical geometry.

Range of Control System

6.31. The range requirements or, in other words, the total reactivity equivalence of various means of control, especially safety rods and back-up safety devices, are fixed almost entirely by design and operating criteria of the reactor. Important factors in establishing reactivity needs are temperature coefficient effects, degree of fuel depletion to be allowed, cumulative fission product (and other) poisoning, isotope production, experiments, etc. These may demand a total of 1 or 2 per cent of additional reactivity (or $\Delta k/k$) for a reactor of low power, or they may need as much as 20 per cent or more for reactors of high flux and high specific power.

Temperature Effect

6.32. Since reactors inherently have (or are designed to have) negative temperature coefficients, the reactivity decreases when the temperature rises, as it inevitably will when the reactor operates at appreciable power. A reactor which is just critical when cold will thus become subcritical when the temperature increases. A certain amount of excess reactivity, i.e., additional fuel, must therefore be included in the reactor to overcome the negative effect of temperature during operation. In reactors of the Swimming Pool type, for example, in which enriched uranium is the fuel and water is both moderator and coolant, the temperature coefficient of reactivity is about -7.5×10^{-5} per °F. When operating at a power of 100 kw the maximum temperature rise is 50°F, and so the reactivity allowance for temperature changes is $(7.5 \times 10^{-5})(50) = 0.00375$. For a reactor of higher power the temperature increase may be greater; if so, a larger temperature allowance will be necessary (see Table 6.1).

Fuel Depletion

6.33. In order to avoid the necessity for shutting down the reactor at frequent intervals to replace the fuel that has been consumed, additional fuel, over and above the essential requirements, is included at the time of initial start-up. In the Swimming Pool reactor this has a reactivity equivalent of 0.0015 and is sufficient for the fuel burnt up in 2400 hr of operation at a power of 100 kw. In a reactor having natural (or partially enriched) uranium as fuel there will be a certain amount of conversion of uranium-238 into fissionable plutonium-239; the allowance for fuel depletion, for a given energy output, will then be less than in a reactor using highly enriched material.

Fission Product Poisoning

6.34. The most important fission product poisons are xenon-135 and samarium-149, and the excess reactivity required to override the effects of these isotopes can be estimated from the equations given in § 4.95, *et seq.* Assuming an average flux of 6×10^{11} neutrons/(cm²)(sec) for the Swimming Pool reactor at 100 kw, the xenon poisoning at equilibrium, calculated from equation (4.104.1), is about 0.0045. As seen in § 4.99, this is approximately equal to the corresponding decrease in reactivity and so an excess reactivity of 0.0045 must be included to overcome the xenon poisoning during operation. Because of the relatively low thermal neutron flux the increase in xenon concentration after shut-down is negligible and so requires no reactivity allowance.

6.35. The equilibrium value of samarium poisoning during reactor operation is 0.012 independent of the neutron flux (§ 4.115). However, at a flux of 6×10^{11} neutrons/(cm²)(sec) it would require over four years for the equilibrium concentration of samarium to be attained. The poisoning during 2400 hours of operation may thus be ignored.

6.36. Other fission products also cause some poisoning but the corresponding excess reactivity requirement is small. An approximate estimate can be made from the known yields of the fission products and their absorption cross sections, and this is usually included with the reactivity allowance for fuel depletion.

Isotope Production, Experiments, etc.

6.37. The reactivity requirements considered above are applicable to all reactors. In many cases, however, further allowance is necessary for neutron absorption resulting from the introduction of extraneous materials for isotope production, experiments, etc. Allowance for the neutron losses thus incurred must be made by including additional reactivity, the amount depending on the purpose for which the reactor is intended.

6.38. In order that the reactor may be raised to full power from shut-down in a reasonable time, e.g., about 10 min, it is necessary that some excess reactivity, roughly 0.003, should always be available. A reactor designed for low-power operation will not have much excess reactivity and so it is desirable to add this operational requirement. In a high-power reactor there is so much excess reactivity built into the system that no special additional allowance need be made.

6.39. The reactivity specifications of two thermal reactors using highly enriched fuel, one operating at low power (100 kw) and the other at high power (30,000 kw) are summarized in Table 6.1.* It will be noted that in the high-

TABLE 6.1. REACTIVITY SPECIFICATIONS FOR CONTROL DESIGN

	Reactivity (or $\Delta k/k$) Requirements	
	Low-power Reactor	*High-power Reactor*
Temperature	0.00375	0.008
Fuel depletion	0.0015	0.035
Xenon and samarium poisoning		
Equilibrium	0.0045	0.050
Shut-down override	—	0.048
Other effects (isotope production, etc.)	0.02	0.02
Operation	0.003	—
Total	0.033	0.161

power reactor 0.048 of reactivity is included to allow for override of fission product poisoning after shut-down. This is sufficient to permit the reactor to be started up within about 30 min after shut-down (see Fig. 4.12). Of course, as long as some of the fuel depletion allowance remains, this will also serve to override the poisoning, so that start-up will be possible even after a somewhat longer period of shut-down.

6.40. The total reactivity requirements given in Table 6.1 represent the minimum fuel loadings, but in practice the reactor is designed to permit a larger amount of fuel to be included in the core in order to provide for unforeseen or special circumstances. For example, it may be found possible to operate the low-power reactor at 1000 kw instead of 100 kw, and this will require additional fuel because of the increased rate of burn-up, and larger allowances for temperature and fission product poisoning. The maximum fuel loading of the reactor may thus be 50 per cent more than the amount corresponding to the reactivity requirements in Table 6.1.

6.41. In the design of the control system for a reactor allowance must be made for the possibility that it may have the maximum fuel loading; in addition the control rods should include a margin, generally about 50 per cent, for control flexibility and to act as a safety factor. Thus, the reactivity (or $\Delta k/k$) range which must be covered by the safety rods is at least 0.08 for the low-power reactor and about 0.4 for the one of high power. The actual number of rods used will depend on the power and size of the reactor, but a reactivity equivalent of 0.05 is common for a single safety rod. The low-power reactor will then require two such rods whereas the high-power reactor considered above will need eight rods.

6.42. The reactivity range of the shim rods is also determined largely by the

* Cf. T. E. Cole, *Nucleonics*, **11**, No. 2, 32 (1953).

reactor design; this is especially true, of course, when the same rods are used for coarse control and for safety purposes. Regulating rod range, on the other hand, is left largely to the control designer, since its function is to permit fine operating control. For the reason given in § 6.27 the reactivity equivalent of a regulating rod is kept below 0.0075, a common value being 0.005. As a general rule only one regulating rod of this reactivity would be used, although another might be available as a stand-by.

Effectiveness of Control Rods

6.43. For the purpose of control rod design, it would be desirable to know in advance the effect on the reactivity of a particular control material placed in a specified position in the reactor. Although the theoretical calculation of the reactivity equivalent of a control rod is a difficult problem, it has been solved in a number of special cases.* Most of the expressions obtained are too involved to be given here, and reference will be made only to the simple case of a cylindrical reactor (radius R) with a concentric cylindrical absorbing rod (radius R_0), as shown in Fig. 6.2. By making the calculations on the basis of a single group of (slow) neutrons (§ 3.201, *et seq.*), and applying a correction factor to allow for the fast neutrons, the change Δk in the multiplication factor resulting from the complete insertion of a control rod of radius R_0 in a reactor of radius R is found to be given approximately by

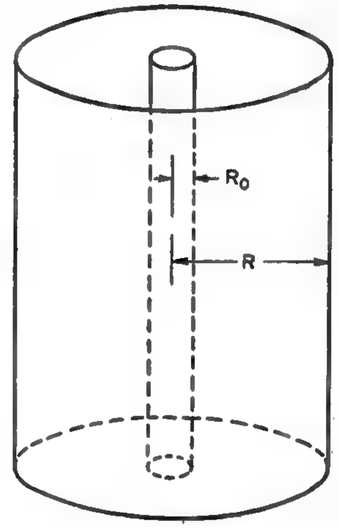


FIG. 6.2. Cylindrical reactor with central control rod

$$\Delta k \approx \frac{7.5L^2}{R^2}\left[2.3 \log \frac{R}{R_0 - d} - 0.76\right]^{-1}, \quad (6.43.1)$$

where L is the diffusion length of slow neutrons in the reactor and d is the linear extrapolation distance at the surface of the rod (§ 3.24). The material of the rod is assumed to be a perfect ("black") absorber for slow neutrons.

6.44. Consider a water-moderated, cylindrical reactor of radius 25 cm, with a concentric control rod of radius 3.0 cm. The diffusion length of thermal neutrons in the reactor is about 2.0 cm and the diffusion coefficient is 0.18 cm, so that the transport mean free path (λ_t) is roughly 0.54 cm. Since the radius of the rod is moderately small, the linear extrapolation distance may be taken as approximately equal to λ_t, so that d is 0.5 cm. Upon substituting these values into equation (6.43.1) it is found that insertion of the rod would change the multiplication factor by about 0.03, i.e., $\Delta k/k$ is roughly 3 per cent. Although

* See, for example, S. Glasstone and M. C. Edlund, "Elements of Nuclear Reactor Theory," D. Van Nostrand Co. Inc., 1952, Chapter XI.

this result is by no means exact, it does give the order of magnitude to be expected.

6.45. Since the neutron flux is a maximum along the central axis of a cylindrical reactor, a concentric rod will exert the maximum effect in reducing the reactivity. The same rod located eccentrically in the reactor will be less effective. Calculations indicate that its effectiveness at any point is approximately proportional to the square of the thermal-neutron flux at that point. Assuming a cosine (or similar) distribution of the flux (§ 3.117), it would appear that a vertical rod located halfway along the radius of the cylindrical, bare reactor would be about half as effective as the same rod at the center. For a reflected reactor, the ratio would be somewhat larger, because of the higher neutron flux away from the center.

Location of Control Rods

6.46. In general, a reactor will have several control rods, and it should be noted in this connection that the over-all effectiveness of two or more rods is not necessarily equal to the sum of the values for the individual rods. The total effect is a function of the spacing between the rods, as well as of their location in the

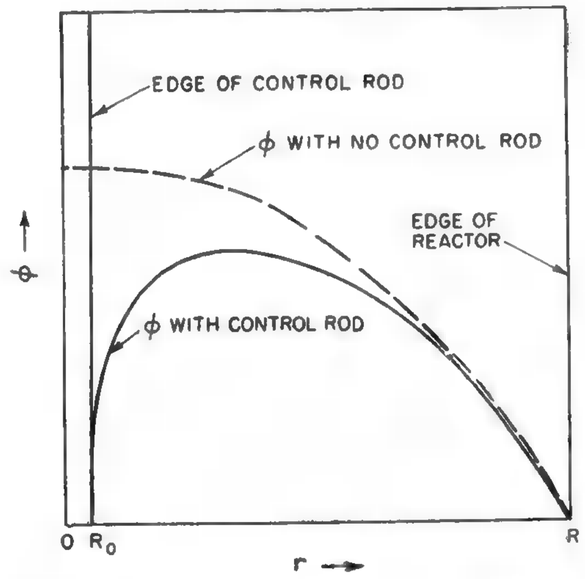


Fig. 6.3. Effect of control rod on neutron flux distribution

reactor. With increasing separation the total effectiveness first increases, passes through a maximum, and then decreases. The explanation of this behavior is that the introduction of a control rod into a reactor distorts the flux distribution; this is shown qualitatively in Fig. 6.3. for a cylindrical bare reactor with a central control rod. It is seen that near the rod the flux is greatly decreased, but at some distance away the proportionate decrease is much less. If a second rod

is inserted at a point where the flux is highest, it will be much more effective than if placed either closer to or further from the first rod.

6.47. In deciding upon the location of the control rods in a reactor, the important point to bear in mind is the desirability of maintaining a fairly uniform neutron flux distribution, and hence of energy release, in order to maximize the power output (see Chapter XI). For this reason, control rods — especially shim rods, since their reactivity equivalent is large — should be located symmetrically. Further, they should be inserted or withdrawn in such a manner as to retain, as far as possible, the roughly uniform flux distribution. The location of regulating rods is less important, since they effect the reactivity, and hence the flux, to a relatively small extent.

6.48. If a thermal reactor has a good reflector, e.g., of beryllium, the neutron flux in the reflector near the core is high. Consequently good control is possible by means of absorbing rods, containing cadmium or boron, located in the reflector. In the Materials Testing Reactor (MTR), for example, the regulating rods are situated within the beryllium reflector. It is of interest to note in this connection that when the Los Alamos Water Boiler was redesigned and the beryllium reflector was replaced by one of graphite, the regulating rods, which had previously been in the reflector, were transferred to re-entrant thimbles in the core vessel. The reason for this change is that graphite is a less efficient reflector than beryllium; the thermal-neutron flux in the former is thus relatively smaller and control by an absorber in the reflector is then not so effective.

Calibration of Control Rods*

6.49. Although a rough idea of the reactivity equivalent of a control rod may be obtained by calculation, as indicated above, the values must be checked by direct measurement. In any event, in addition to knowing the total effectiveness of the rod when fully inserted, it is necessary to calibrate it, so that its reactivity equivalent is known for various degrees of insertion. The required data can be obtained only by direct measurement on the reactor itself or, preferably, on a low-power mock up or critical assembly.

6.50. One method used is the following.† The rod to be calibrated is completely withdrawn, and the experimental assembly is made slightly supercritical, so that the neutron flux is increasing. The reactor period is then determined by observation of the rate of increase, e.g., by means of a period meter. The rod is then inserted to a known distance and the stable period is redetermined, after allowing sufficient time for the transient behavior to die out (§ 4.19, *et seq.*). This procedure is repeated at several points, until the rod is completely inserted.

6.51. From equation (4.35.1) and the known delayed-neutron characteristics

* J. D. Richards, Report AECD-3388.

† Another method is based on measurement of the amplitude of the neutron flux oscillations accompanying the sudden insertion and withdrawal of the control rod (see A. M. Weinberg and H. C. Schweinler, *Phys. Rev.*, **74**, 851 (1948); J. I. Hoover, *et al.*, *ibid.*, **74**, 864 (1951)).

(Table 4.1), it is possible to express the relationship between the reactivity, ρ, and the stable period, T, for a particular value of l, the neutron lifetime in the reactor. Thus, if uranium-235 is the fissionable material,

$$\rho = \frac{l}{Tk_{\text{eff}}} + \frac{5.30 \times 10^{-4}}{0.62 + T} + \frac{5.30 \times 10^{-3}}{2.20 + T} + \frac{0.0138}{6.48 + T} + \frac{0.0526}{31.7 + T} + \frac{0.0200}{80.0 + T},$$

with T in seconds; the neutron lifetime may be calculated by the method described in § 3.164. Consequently, from a measurement of the reactor period, the corresponding reactivity can be found; as a first approximation, k_{eff} may be taken as unity. If T is large, as it usually is in calibration measurements, the first term of the equation may be neglected, so that the values of l and k_{eff} need not be known. From the observed reactivities with the control rod in various positions, the reactivity equivalent of the rod can be obtained for different amounts of insertion.*

6.52. The general nature of the results obtained in the calibration of a control rod is indicated by Fig. 6.4; the change in reactivity is shown as a function of the distance the rod is inserted. The S-shaped curve is quite characteristic for

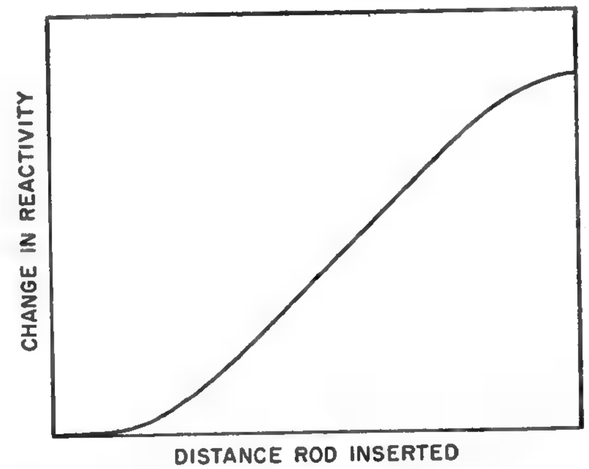


Fig. 6.4. Change in reactivity as function of insertion of control rod

reactors of most types. When the rod is inserted to a small extent, it will affect the reactivity only slightly. Beyond a certain point, the change in reactivity is linear, that is to say, it is directly proportional to the extent of insertion of the rod. Finally, when the rod is almost completely inserted, further insertion has relatively little effect. The small changes in reactivity at the beginning and end of the rod insertion can be accounted for by the lower neutron flux near the edges of the reactor as compared with that closer to the center.

* The reactivity can be expressed in terms of the inhour unit, but this is not recommended for general use (see § 4.43).

Accuracy of Control System

6.53. Whereas range is a matter involving only the control rods, accuracy concerns the whole control system. Accuracy of safety action, in the sense of promptness and reliability, must be specified in particular relation to the shortest period of which the reactor is considered capable (see § 6.116). The increased speed of response must not be obtained, however, at the expense of stability or reliability. To meet such a requirement may call for detailed attention to instruments, control block, and rod release and action mechanisms.

6.54. In this connection, it should be noted that the control system should function as rapidly as possible to provide the best protection against the effect of transients. In general, the heat capacity or mechanical design of a reactor will be such that it can be overloaded to a certain extent for a short length of time without suffering damage. In other words, a particular amount of energy overload is permissible before some portions of the reactor would be distorted or destroyed. This amount of energy then effectively defines the speed of the control system necessary to deal with the largest expected transient. If it is possible to increase the speed of response of the control system over that required by the thermal properties and the mechanical design of the reactor, then additional safety will be provided. It should be understood, of course, that stability and reliability of components must be maintained. Frequently, however, reliability decreases as the speed of response of the system increases; hence, as in other engineering fields, a compromise must be made.

Efficiency of Control System

6.55. The specification of efficiency is largely independent of reactor design. It refers, essentially, to quality of control, indicating the extent to which a control system, meeting other, more fundamental, specifications, has been achieved at a low cost in wasted neutrons, in power consumption, and in disturbance or restraint of operation. It is not surprising that, in the brief history of reactor design, effectiveness of the control system has received more attention than efficiency, at least as far as neutron economy is concerned. Use of boron-steel shot, for example, must be rated as inefficient since it is not an easy matter to remove the shot from the reactor. Nevertheless, it provides a simple and effective back-up safety device. In general, more efficient control methods, in the sense referred to above, are to be expected in due course.

6.56. The problem of avoiding disturbance or restraint of operation, as far as possible, has, however, received considerable attention. In this connection four general principles have emerged as being basic to control design. These are (1) the principle of duality, (2) the fail-safe principle, (3) the sequential principle, and (4) the principle of reliability.*

6.57. The duality principle requires that essential services should be in du-

* I. A. M. Garcia, Report AECD-3591.

plicate. For example, two complete and, if practical, entirely independent channels of power feed are desirable, a normal feeder and an alternative to be available in the event of an emergency. For each critical control function using relays or contactors a duplicate should be included to guard against failure.

6.58. In the event of a serious power, instrumental, or operator failure the control system should act rapidly to shut down the reactor. But this is not the only aspect of the fail-safe principle. It is also important that a minor failure should not interrupt reactor operation. This result can be partly achieved by duplication, as stated above, and also by the use of devices which do not give misleading indications when they fail. Attention of the operator should be called to equipment failure by a suitable annunciator, e.g., a light, a drop indicator, or an alarm, so that the faulty device can be replaced immediately.

6.59. The sequential principle in control design means that the various operations necessary for start-up and for normal or emergency shut-down must take place in a specified order. To attain this end a number of interlocks are incorporated in the control system. In start-up it should be made impossible, for example, to withdraw a shim or regulating rod unless the safety rods have been raised, at least partially, so that they can be dropped in an emergency. On the other hand, insertion of a shim or regulating rod should be possible at all times. Provisions may be available for by-passing the operational sequence if necessary, but adequate safeguards must be included to allow for human or other error.

6.60. Finally, failures can be minimized by using the most reliable equipment. In some instances, where sufficiently reliable devices are not available commercially, special pieces of equipment may have to be made. Some engineers prefer not to use vacuum tubes where reliability is the prime consideration. Nevertheless, these tubes have so many advantages that they are widely employed in reactor control, with adequate precautions against failure. Another aspect of reliability is circuit reliability. This implies, in general, simplicity, but this is, unfortunately, not always compatible with safety. The latter requirement, which is particularly severe in reactor operation, inevitably introduces some complications.

Basic Information for Control Design

6.61. In summarizing the foregoing discussion, it may be repeated that complete design of a control system must be based on information concerning reactor design and reactor operation. Some typical items involved are recorded in Table 6.2. Several of these are basic to the entire reactor design and are influenced by many considerations not relevant to control. It is to be expected that, in some details of the items in the table, changes may be made as the overall reactor design, including control design, progresses. Actually it is desirable that the control designer should take part in the preliminary design phases of

TABLE 6.2. INFORMATION BASIC TO CONTROL DESIGN

Reactor Design

Total reactivity needed for temperature, poisoning, fuel depletion, etc.
Method of control chosen, e.g., motion of absorber, moderator, reflector, or fuel.
Number and location of control "rods."
Transients, e.g., reactivity, thermal.
Special hazards, e.g., leakage of coolant, disintegration of fuel elements.

Reactor Operation

Purpose of reactor, e.g., criteria of good control performance.
Operating routine, e.g., frequency and duration of shut-downs.
Maintenance and servicing routine.

the whole reactor system in order that advantage may be taken of such inherent features as make for ease of control.

6.62. The task of the control designer is to take the basic information, as outlined in Table 6.2, and to proceed toward his own objective, namely, to design an operator loop and an automatic loop, so coordinated as to give the desired quality of operation, with as much safety as is reasonably possible. To achieve these ends, it is necessary to design or specify various devices and equipment, including instruments, effectors, control computer, and control room, which will permit effective and safe operation of the reactor.

THE INSTRUMENT BLOCK *

Choice of Control Quantities

6.63. An important part of the problem of designing the instrumentation system for any reactor is the choice of those particular instrument signals upon which control action should be based. The selection will in all cases be governed to some extent by the purpose of the reactor under consideration and by the relative emphasis laid upon safety and on operating control. However, as in other aspects of reactor design, there are certain basic principles of wide applicability.

6.64. It is perhaps appropriate to begin by noting the uses to which the various instrument signals may be put. Bearing in mind that attention is here confined to instrumentation relevant to operation and control, these uses may be listed as follows:

1. Quantitative indication, e.g., of power or its equivalent, which is the normal function of indicating instruments.
2. Alarm indication, involving a special visual or auditory indication.
3. Automatic operating control.
4. Automatic safety action.

* For general reviews, see J. M. Harrer, *Nucleonics*, **11**, No. 6, 35 (1953); J. D. Trimmer and W. H. Jordan, *loc. cit.*

5. Recording, most of which is not for the immediate purpose of operation or control.

6.65. To the extent that the operator is in control, all information presented can conceivably enter into control of the reactor. Practical design must concentrate on giving the operator all the information needed to operate — smoothly in the usual situations and effectively in the unusual ones. Some limitations on elaboration of instrumentation for operator control are expense and the possibility of providing so much information as to be burdensome or confusing. In any event, it is desirable that the operator should be bolstered by automatic safety devices to take care of accidental or unexpected situations.

6.66. When the reactor is functioning under automatic control, it is probable that one or two instrument signals will be in full control at any given time. It is here, therefore, that the choice of a suitable control quantity is especially important. It is a reasonable generalization to state that in research or isotope-producing reactors, where neutron flux is of direct significance, some measure of neutron level is the logical choice. In power reactors, on the other hand, where over-all power output is more significant, coolant flow and coolant temperature may sometimes take precedence. Nevertheless, even in such reactors, the neutron flux signal has certain advantages, notably in its high speed of response, which would recommend its inclusion in the control formula. The safety system may well involve a suitable combination of power and neutron signals, as will be seen in Table 6.4.

Choice of Control Instruments

6.67. The indicating instruments in most general use for various aspects of reactor control and operation, based on measurements of neutron flux, are listed in Table 6.3.* A simplified block diagram of the instrumentation of a reactor,

TABLE 6.3. REACTOR INSTRUMENTATION: NEUTRON DETECTORS

Purpose	*Detector*	*Indicating Meter*
Power indication (Counter range)	Fission (ion) chamber or BF_3 proportional counter	Counting-rate meter (logarithmic scale)
Power indication (Period range)	Compensated ion chamber	Log n meter
Period indication (Period range)	Compensated ion chamber	Period meter
Power indication (Power range)	Compensated ion chamber	Galvanometer or Micromicroammeter
Automatic control (Operating level)	Parallel-plate (PCP) chamber	(To servomechanism)
Power indication (Operating level)	Boron thermopile	Voltage indicator

* Cf. J. R. Huffman, *Nucleonics*, **12**, No. 4, 21 (1954).

which may be taken as fairly typical, is also shown in Fig. 6.5. The safety system is not included, as this will be considered later (§ 6.96, *et seq.*). The detecting instruments mentioned and the associated meters have been described in Chapter V. The detectors are, of course, placed in the reactor, either in the core or in the reflector, and the meters are on a desk or on panels in the control room. Some of the electronic equipment, such as the preamplifier, is usually near the reactor, and the remainder is at the control center. In addition to the

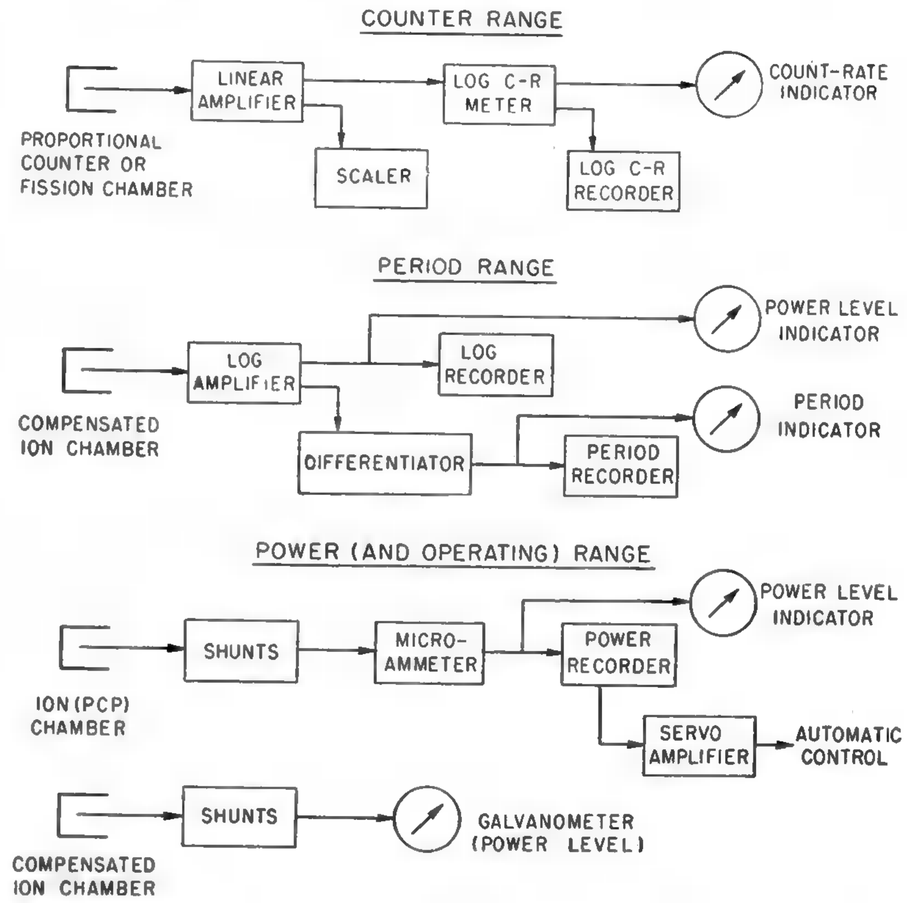


FIG. 6.5. Block diagram of reactor instrumentation for start-up and operation

instruments mentioned in Table 6.3, others are often used for power determination involving measurements of the temperature increase of the coolant and its rate of flow (cf. § 5.128).

6.68. Mention may be made of the important matter of locating the neutron detecting devices in the reactor. Apart from considerations of instrument sensitivity and the effects of radiation on the materials (§ 5.116), the location of a detector with respect to a control rod may have a significant bearing on the interpretation of the signal obtained. This is especially the case if the particular

instrument signal plays a direct part in controlling the motion of a regulating rod. In general, the problem of instrument location, like that of locating the control rods (§ 6.46), requires a qualitative understanding, at least, of the way in which movement of a rod will affect the local flux distribution.

Rod Position Indicators

6.69. It is important that the operator should know the extent of insertion of the various control rods into the reactor at all times. For this purpose rod position indicators are employed. A simple form of such a device is indicated diagrammatically in Fig. 6.6. When the rod is moved the rotation of the pinion wheel, of the rack and pinion system which drives the rod, causes a rotation of the attached synchrogenerator. The output of the latter energizes the field of a synchromotor located in the control room. This causes an indicator to move by an amount corresponding to the motion of the control rod. In order to avoid any ambiguity concerning the position of a rod it is the general practice to have the synchrogenerator so geared that the indicator makes less than one

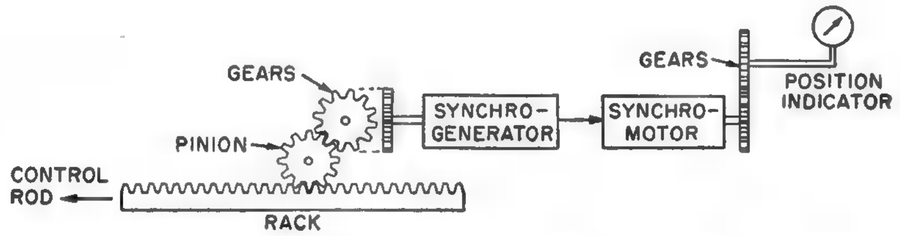


Fig. 6.6. Diagrammatic representation of control-rod position indicator

revolution for full travel of the rod. For more precise indication, a second synchrogenerator making several revolutions may be used or the signal may be amplified to energize a motor which drives a synchrocontrol transformer. This then moves dials in the control room indicating the position of the rod.

6.70. Besides the instruments which indicate rod motion and position, there are frequently lights on the control desk connected to limit switches. These lights come on when a rod reaches the limit of its motion either in or out of the reactor. The attention of the operator is thus specifically drawn to a particular condition which might require immediate action.

THE OPERATOR AND CONTROL BLOCKS

Principal Elements and Functions

6.71. In the general block diagram of the control system (Fig. 6.1), the control block is shown between the operator and reactor blocks in the operator loop, and between the instruments and the reactor in the automatic loop. It is the purpose of this section to give consideration to communication among

instruments, operator, and control blocks, and to the elements and functions of the control block itself.

6.72. The functions of the operator and of the control block are fundamentally the same. As stated earlier, the reactor could conceivably be controlled completely by the operator, even to the extent of moving the control rods by hand. These common functions are: (1) to accept information from instruments, (2) to remember information, (3) to reach decisions, and (4) to exert control action. For example, an operator might notice a signal from an instrument telling him that it would be wise to shut the reactor down. He might feel some doubt, however, as to the reliability of this information, and decide to wait. If soon thereafter a second, independent instrument signaled the advisability of a shut-down, the operator, remembering the first signal, would probably reach a decision in favor of action, and would proceed to take that action. An analogous sequence of events could occur entirely within the automatic loop. This could be designed so that it would initiate shut-down only upon the concurrence, within some specified time, of two or more independent instrument signals.

6.73. In principle, therefore, the operator can do anything the control block can do. There are, nevertheless, excellent reasons for having both. For one thing, movement of the control rods may be beyond the strength of the operator, so that it is necessary to have a motor to do the work. Further, the reaction time of the operator sets a limit upon the speed with which he can accept information and take action. Consequently it is advisable to support the operator by an automatic safety system which will always be ready for high-speed, positive action.

6.74. Since automatic control appears essential, at least to some extent, it may be wondered why an operator is needed. The answer is that an automatic system is subject to mechanical failure and it can, in any event, be no better than the limited decisions built into it by the designer. Circumstances might thus arise in which an on-the-scene operator has information available which could not possibly have been predicted by the control designer. It would appear, therefore, that in controlling reactors, especially those which are to be flexible and capable of being used for a variety of types and conditions of operations, the human operator will always have an important and irreplaceable role. In fixed-purpose reactors, with rigid operating routines, however, there is scope for a fuller degree of automatic control.

Function of the Operator

6.75. The human operator is valuable as part of a control system because of the flexibility and variety of his responses to different situations. This variety makes it difficult to describe his functions completely, but, in general, it may be said that the operator's activities fall into two main groups, namely, routine and emergency. Under routine activities would be included all operations to be expected for the normal functioning of the reactor, such as start-up, shut-down,

change of power level, and operation at fixed level. Emergency operations, which might be regarded as operation in face of difficulty, would involve such situations as power failure, instrument failure, automatic shut-down action, change from one automatic control channel to another, and unintentional loss of power level with consequent increase of xenon poisoning.

6.76. A good control system design should satisfy the following three conditions which are particularly relevant to the operation:

(1) *In routine operation* the role of the operator can be minor and straightforward, with as much of the operation as is permitted by cost and other limitations being automatic.

(2) *For emergency operation* the versatility of the operator is valuable and should be given as much scope as is consistent with safety.

(3) *At all phases of operation* positive, safe shut-down action should be accessible both to the operator and to the automatic safety circuit.

6.77. In a complex reactor, having a large number of instruments and involving many interrelated operating restrictions, a device called a *procedure panel* has been found useful, both to the engineer in the preliminary design phases and to the individual engaged in reactor operation. It is equipped with a series of lights which come on successively as the conditions are satisfied for running the motors operating the various control rods. The purpose of the procedure panel is to help the operator in checking and remembering the status of the reactor, especially during start-up and shut-down.

Transmitting and Receiving Information

6.78. The human operator of a reactor works at a control desk or console upon which are mounted indicating instruments and manual switches (see § 6.83). Around the console are grouped other instruments and devices, usually mounted on upright panels. A telephone is available whereby the operator may receive information and transmit orders. The entire control room set-up is designed to put the operator in a position to receive information which may arrive by way of counters, meters, recorders, annunciators, etc. These devices will tell the operator the status of the reactor at all times and warn him of the failure of any piece of equipment.

6.79. As a result of the information received in this manner, the operator can formulate decisions and can take action putting the decisions into effect. The operator is consequently part of a two-way communication chain. Information comes to him from instruments and possibly by telephone, and, after making a prompt decision, he passes on information in the form of orders by pushing buttons, turning handles, or speaking into a telephone. Thus control-room design is, in part, a problem in communications.

6.80. The foregoing remarks, concerning the position of the operator in the operator loop, apply in large measure to the position of the control block in the automatic loop. The control block, like the operator, receives information from

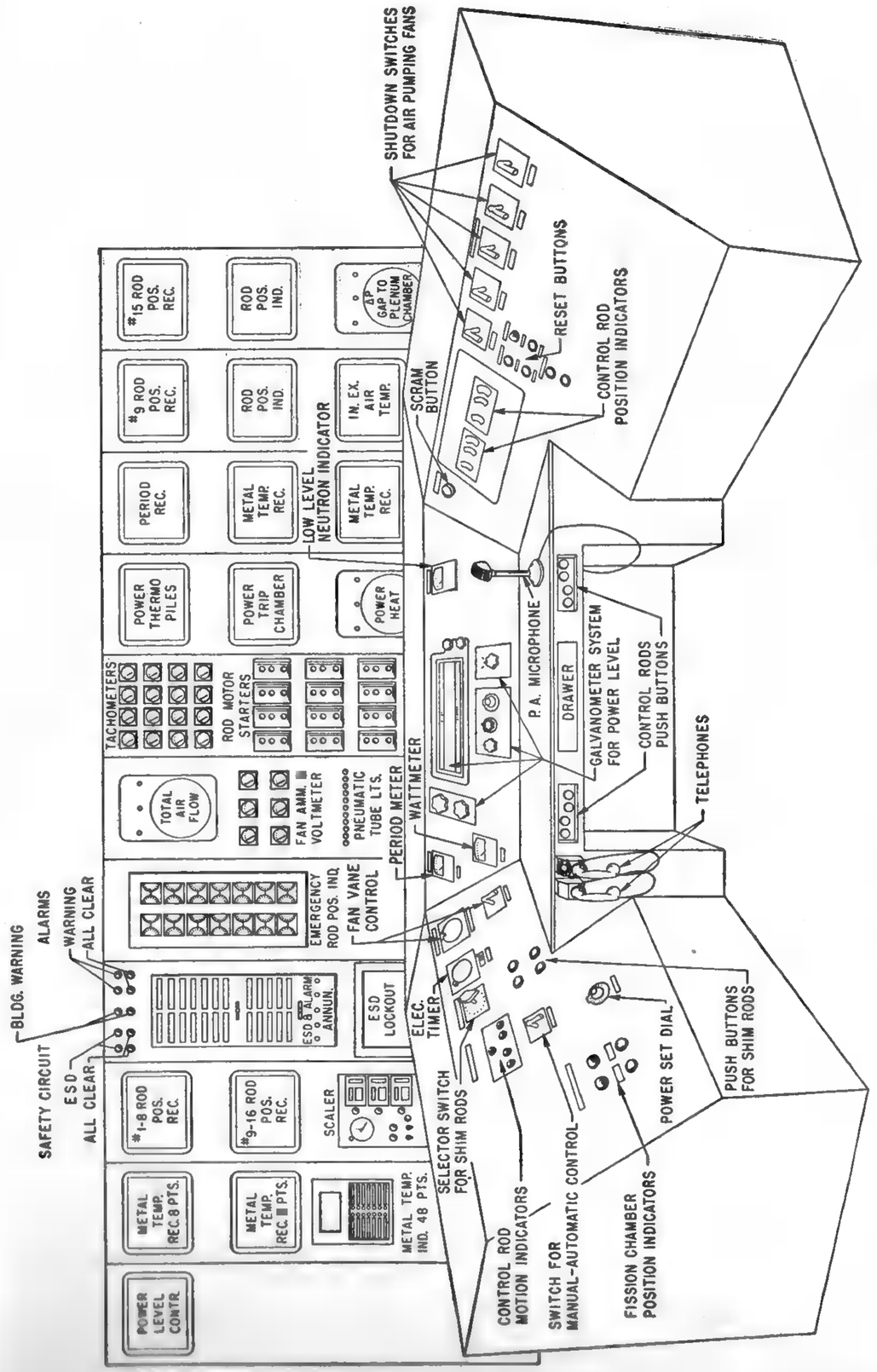


FIG. 6.7. Control console and panels of Brookhaven reactor

operator and instruments, formulates decision, and puts orders into effect. There are, however, two important differences between the situation of the control block and that of the operator. First, is the fact that the operator affects the reactor through the control block, whereas the latter acts directly on the reactor. The control block, therefore, includes motors or, in general, "effectors" which do the work of actually moving the control elements in the reactor. The second difference is that, in contrast to the operator's decision, over which the control designer cannot exercise authority, the decisions put into effect by the control block are determined solely by the control designer. The decisions built into the control block are based on data concerning reactor design and operating criteria discussed earlier (cf. Table 6.2); in addition, safeguards must be included to take care of certain predetermined situations which might be hazardous. The means of building the decisions into the block are discussed below.

6.81. The problem of conveying information from instruments to control block and operator is not appreciably different for reactor control than for any other process control. The laying out and connecting up of the requisite electric, hydraulic, pneumatic, or mechanical lines must be done with great care. The only feature peculiar to reactor control is the neutron flux or other radiation to which the lines may be subjected. Some aspects of this problem, as far as neutron-detecting instruments are concerned, have been mentioned in Chapter V.

6.82. Design of the control console and other control-room features is also much the same for reactor operation as for other processes, such as electric power generation and industrial chemical processes. Due consideration must be given, however, to the degree of continuing alertness and attention which the operator must give to his task. This varies considerably from one reactor to another, but in general it is greater in reactor operation than in more conventional process controls.

6.83. A good example of the layout of a reactor control room is shown in Fig. 6.7, which is a representation of the control console and panels used in operating the Brookhaven reactor.* On the console are located the main control switches and buttons which permit the operator to start and stop the motors that move control rods; to change from one recording instrument to another; to transfer from manual to automatic control; to scram the reactor, etc. The most important indicating meters are also on the console; they show the neutron flux (or power level) and reactor period during start-up and shut-down and also the operating power level. An instrument for indicating the deviation of the actual power level from the desired value may be included. Other instruments, particularly those showing the positions of the control rods, and coolant and fuel element temperatures, are on the panels where they can be seen by the

* Private communication from M. Fox.

operator seated at the controls. Instruments which provide permanent records of the operating behavior of the reactor are also located on these panels.

Control Effectors

6.84. As noted above, the control block must include the motors and other effectors capable of actually exerting the control action on the reactor. The effectors may take orders from operator signals, from instrument signals, or from a combination of both. The type of effector used will depend on the particular function it has to fulfill.

6.85. Consider, for example, the effectors required for moving the various kinds of control rods. The most common effectors for shim and regulating rods are electric and hydraulic motors. A convenient way in which the motion may be accomplished is indicated in Fig. 6.8; this is a feedback, closed loop system for altering the position of a control rod.* The switch handle, located on the

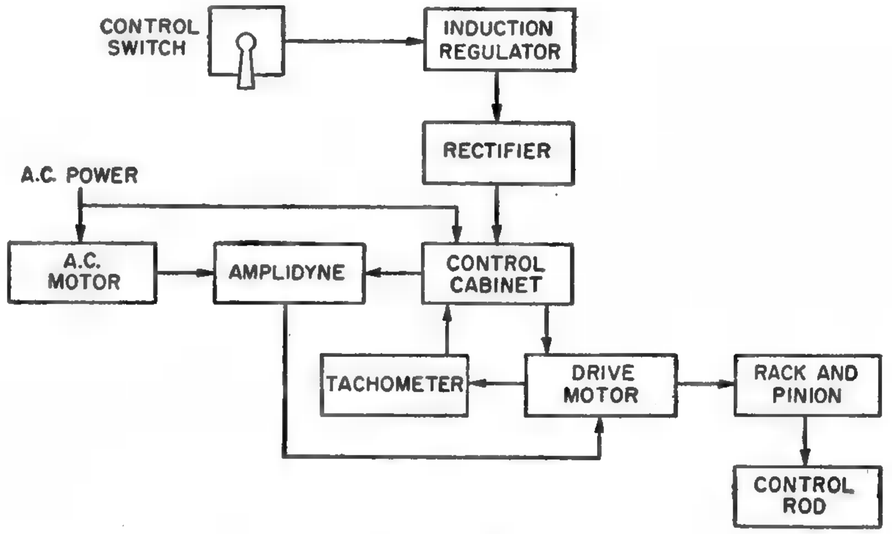


Fig. 6.8. Block diagram of rod-motion system

control console, is coupled directly to an induction regulator, and when the handle is moved a current is induced in the latter. The output is rectified and the voltage produced across a resistor is balanced against the voltage from a tachometer generator connected to the motor which drives the control rod through a rack and pinion system. The difference between the two voltages is applied to the control field of an amplidyne generator, driven at a constant speed by an a-c motor. The amplidyne output is then fed directly to the driving motor armature. This motor has a fixed field and so the speed is proportional to the displacement of the control handle. Thus the rate of motion of the con-

* I. A. M. Garcia, *op. cit.*

trol rod is also proportional to the latter. The clockwise or anticlockwise rotation of the control handle determines the direction in which the rod moves.

6.86. For safety rod motion, which must be rapid and positive, it is the general practice to call on a store of energy, such as gravity or a tank of fluid at high (hydrostatic) pressure (Fig. 6.9).* The corresponding effectors, which would be released in the event of an emergency, would be a lifting magnet in the former case or a valve in the latter case. Since the gravitational store of energy is perhaps the most reliable, there is an advantage in having vertical shim rods which can also be used as gravity-actuated safety rods (cf. § 6.29). Horizontal

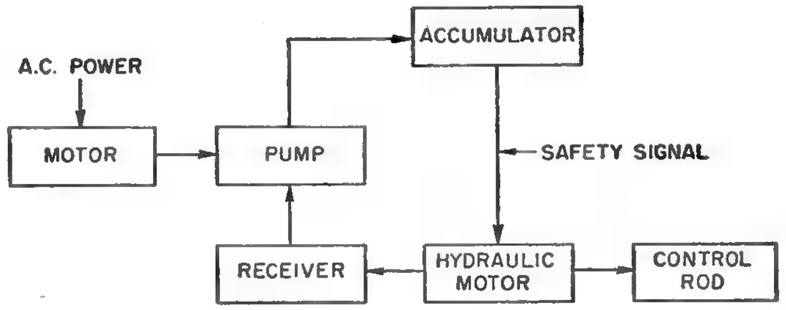


FIG. 6.9. Block diagram of safety system using hydrostatic pressure

operation, as in the Brookhaven reactor, however, puts a more symmetrical load on the effectors. Since gravity cannot then be used, the safety rods are operated by hydrostatic pressure. For back-up or emergency safety devices, such as dropping boron-steel shot, release of a fluid, etc., the effector is either a manual or electromechanical relay, catch, or valve.

6.87. A problem which has a direct bearing on effector specifications is the location with respect to various radiation and pressure shields around a reactor. Control rods are located in the core or in the reflector, and so the effectors may have to operate under adverse conditions of nuclear radiation, temperature, and corrosion; they may also have to operate control elements through a pressure seal, e.g., in a pressurized-water system. The presence of seals is particularly objectionable for safety rods, because of the danger of sticking. One device for introducing control rods (and instruments) to the immediate vicinity of the core without going through pressure seals is the use of thimbles welded into the containing vessel, as in recent forms of the Water Boiler reactor.

6.88. In general, effectors may be grouped into three classes; these may be described, in brief, as (1) latches, (2) motors, and (3) servomechanisms. As already indicated, the typical use of latches, including valves, clutches, etc., is in regular and back-up safeties. The term "motors" is used here to typify any effectors needed for coarse regulating action, such as motion of shim rods or changing the level of a liquid moderator, which cause a relatively slow rate of

* I. A. M. Garcia, *op. cit.*

change of reactivity. Servomechanisms refer to the devices which are used for automatic movement of regulating rods, for high-speed, fine control actions. Each of these three classes of effectors has been the subject of considerable study, apart from their application in reactors. Except for instances in which the special conditions mentioned earlier apply, the requirements for reactor control are not fundamentally distinctive. It is probable, however, that in the interest of automatic operation and relieving strain upon the human operator, reactor development will make increasingly strong demands on the design of servomechanisms.

Incorporation of Decisions into the Control Block

6.89. When the control designer has developed a complete picture of how the operator loop and the automatic loop should function, he must decide on the actual hardware to go into the control block to permit this functioning. The hardware falls into two distinct, but not necessarily separate, categories: that which serves to put into effect as control action the decisions of the operator, and that representing the decisions built into the controls. The combination of these two aspects may be called the "control computer."

6.90. Automatic control of period and of power level, by means of a servomechanism, and the various circumstances which lead to automatic safety actions are examples of the kind of decisions which must be implemented by suitable hardware in the control block. These matters will be considered in later sections. Some reference will be made here to the interlock circuits whose purpose is to exclude, to a large extent, the possibility of human error and of misoperation of the reactor.

6.91. Unlimited addition of interlocks could, no doubt, provide against all foreseeable contingencies, but the control system would then be so complicated as to be impractical. Consequently in the design of interlocking circuits the controls engineer must strike a balance. While possibilities of serious potential hazard must be removed from human control, the system must be flexible enough to permit the operator to use his discretion in certain circumstances.

6.92. Some of the decisions which may be incorporated in the control block are given below; brief comments on the bases for these decisions are included, although in most cases they are readily apparent.

(1) At start-up no control rod can be withdrawn until the counting-rate meter shows a reading. This insures that the reactor is at least in the counter range and minimizes the danger that the neutron flux will increase too rapidly without the operator being aware of the fact.

(2) Safety rods cannot be withdrawn at start-up unless the shim rods (if independent) and regulating rods are fully inserted. This provision is made in an attempt to insure proper sequential action in start-up and to decrease the chances of reaching criticality unexpectedly.

(3) The shim rods (if independent) cannot be moved until the safety rods

have been withdrawn to an appreciable extent. If the shim rods also function as safety rods (§ 6.29), then this provision applies to the regulating rods. Its purpose is to make certain that sufficient safety action is available in the event of an emergency.

(4) When the safety rods can be lowered at two different speeds (see § 6.106) fast lowering, i.e., the greater emergency, must always be able to override slow lowering, i.e., the lesser emergency. Further, lowering of rods must always override raising, i.e., a signal requiring a decrease of power must be given preference over a signal asking for an increase of power level.

(5) The safety (scram) signal cannot be overridden by any action of the operator and it must be impossible to restart the reactor until the condition causing the scram has been rectified.

6.93. The hardware used to translate these decisions into practice are chiefly relay-contactors and manual switches. The relay-contactor consists of a coil, through which a fixed electrical current is either flowing or is not flowing, and of one or more contactors, each of which is correspondingly closed or not closed, i.e., closed or open. A switch is similar, except that manual action replaces coil action. The design of circuits employing these devices is a well defined field of electrical engineering. Since the subject is not peculair to nuclear reactors it will not be treated here and the reader is referred to special works dealing with switching circuits.*

6.94. It is of interest to note that a set of contactors constitutes a "memory." Like any binary device, i.e., one having two, and only two, states, a contactor can store one unit of information. Any binary digital computer consists of an assembly of binary devices, together with means of inducing transitions in these devices from one of the two states to the other. In the interlock circuit system the binary device is the contactor, and the transitions are induced by a relay coil or manual switch; it is thus a kind of digital computer.

6.95. Not all aspects of control computers, however, are of the digital (or relay) type; many are of the analogue or continuous type. An example of a continuous computer is the input circuit to the servomechanism for the automatic adjustment of reactor period and of the operating power level (§ 6.122, *et seq.*). Another instance, of an entirely different type, is the so-called "auctioneer" circuit used in the safety system (§ 6.111).

Design of Safety System†

6.96. The interlock circuits, which permit particular action in certain circumstances only, are, of course, part of the over-all safety system. The present section will deal more specifically with those aspects of the system designed to take action in emergencies. It should be understood that although the purpose

* See, for example, W. Keister, A. E. Ritchie, and S. H. Washburn, "The Design of Switching Circuits," D. Van Nostrand Co., Inc., 1951.

† Cf. J. I. Owens and J. H. Piggott, *Elec. Eng.*, **74**, 306 (1955).

of the safety system is to provide automatic action, the operator must always be able to decrease the power of the reactor or to shut it down completely, by manual action, in any circumstances. In other words, as regards taking safety action, the operator must always be able to override the automatic circuits.

6.97. The safety system is a fundamental part of the control block concerned with both automatic and operator loops. The primary hazards of reactor operation, which must be taken into consideration in designing the safety system, are (1) excessively high level of neutron flux, (2) excessively short period of flux increase, and (3) diminished flow of coolant. Other, secondary, dangers are instrument or equipment failure, confusion or mistakes of the operator, accidents in the assembly, maintenance, or operation of the reactor, and accidents in experiments carried out in the reactor. A safety system is best developed from a pattern laid down by the three primary hazards, with protection against the secondary ones being worked in as far as possible.

6.98. Although there is no such thing as too much safety, practical considerations limit the complexities of safety systems. The most important limitation arises from the basic conflict of safety and operation, for any operation of a reactor, as of innumerable devices in daily use, involves some hazard. In practice, therefore, the safety devices represent a compromise determined largely by the purpose for which the reactor is intended. In operating the reactor of a mobile power plant, such as on a military aircraft, for example, it may become urgently necessary to take certain calculated risks. The introduction of too many safety precautions might cause the reactor to be shut down so frequently as to make its operation virtually useless. Consequently there must always be a balance between safety and continuity of reactor operation.

6.99. Another limitation in safety design is the cumulative probability of trouble or failure in the system as it becomes more and more complicated. This leads to the dilemma of either operating with a partially incomplete, and hence somewhat uncertain, safety system, or of tolerating loss of operating time needed for repair of the system. Finally, mention may be made of the economic aspect of the problem; with safety, as with other commodities, there comes a point of diminishing returns.

6.100. One of the great uncertainties in safety system design is the relative confidence to be placed in the operator and in automatic action. As far as initiation of safety actions is concerned, the difficulty is resolved by permitting both manual and automatic action. There remain, however, such problems as what the automatic system can do to compensate for the operator's mistakes, and what the operator can do about equipment failure. A satisfactory solution to the latter appears to be to keep ultimate safety actions, i.e., scram and back-up, always operative, and to provide "test blocks" which permit certain instruments to be isolated from the safety system and be removed for repair or replacement, without causing the reactor to be shut down. This would allow temporary operation of the reactor without full instrumentation, but never

without safety protection. Those channels on which test blocks have been pulled to permit instrument repair should be brought to the operator's attention by an annunciator.

6.101. A safety action may be more or less severe, both as to the extent and speed with which it affects the reactor, and as to how much trouble it is to recover from it and resume operation. The following actions are listed in order of increasing severity in one or both senses:

1. *Period limit:* prevents increase in reactivity upon reaching a predetermined period, i.e., rate of neutron flux increase, during start-up.
2. *Level (flux or power) limit:* prevents increase of neutron flux upon reaching a predetermined power (or flux) level.
3. *Reverse (or setback)*: automatic slow decrease of reactivity.
4. *Scram:* automatic rapid decrease of reactivity.
5. *Back-up:* emergency clinching of low reactivity.

The first three of these safety actions act as limitations upon the ability of the operator to increase the reactivity when the period or power level (or other situation) is outside the normal range. They are designed to minimize shut-down of the reactor due to operator errors as well as to provide an additional margin of safety.

6.102. Period limitation is most important when the neutron level is low compared to the normal operating level. Because of small excess reactivity and long neutron lifetime, dangerously short periods are not possible with some reactors, but where such short periods can arise (see § 6.116) automatic period protection is essential. Power level limits become increasingly important as the operating level of the reactor is approached, and safety action initiated by a neutron flux slightly above the normal operating level is mandatory.

6.103. In addition to the safety actions based on reactivity or neutron flux it is usually desirable that coolant flow and temperature signals be incorporated in the safety system. Any decrease in the rate of flow or an undue rise in the temperature of the coolant should call for suitable action.

6.104. The secondary hazards of maintenance and operation of the reactor cannot be summarized in a general way, since they vary from one reactor to another. However, mention may be made of rupture of a fuel element leading to escape of radioactive fission products into the coolant, excessive radiation levels of the surroundings of the reactor, power failures, etc. The safety system should permit the shut-down of the reactor, either completely or partially, in such circumstances.

6.105. If every departure from normal operating conditions caused complete shut-down, i.e., scram, the continuous operation of the reactor would become impossible. It is thus desirable to have various degrees of safety action depending upon the extent of the emergency. One way in which this could be done in

a reactor using electromagnetic clutches to support the shim-safety rods and an automatic system for power level control will be described below.

6.106. The exciting current (about 30 milliamp) is supplied by the output stage of an electronic amplifier, called a magnet amplifier, which receives voltage signals indicating reactor period and neutron flux. If the period is shorter than a specified value or the neutron flux exceeds a permitted level the output of the amplifier and, hence, the exciting current of the magnets, is sharply decreased. The safety rods supported by the magnets will then drop rapidly. This behavior is referred to as "fast scram."

6.107. On the other hand, certain signals which do not have the urgency of excessive power level or of a very short period, can cause what is called "slow scram." These safety signals operate relays in the line to the transformer supplying the anode (B+) voltage to the output tubes of the magnet amplifier. When the voltage is interrupted in this manner the exciting current for the magnets decreases relatively slowly because of the time required to operate the relays and to discharge power-supply capacitors, etc. After receiving the scram signal, there is then a short delay before the safety rods are dropped.

6.108. Next in order of emergency is the action known as "reverse"; this is the continuous insertion of the shim rods. During normal operation these rods are stationary, in general, but upon receipt of the appropriate signals the motors driving the rods are started and the rods are inserted. This insertion continues as long as the causative condition endures, but if it is rectified, either spontaneously or by operator action, the rod motion ceases.

6.109. Finally, a reactor safety system may be designed to include the possibility of "setback." This refers to a lower adjustment of the "set point" which determines the power level setting of the automatic control system. When the situation calls for setback, a motor automatically lowers the set point representing the power demand. When the motor runs at high speed, it is called "fast setback"; if the speed is not so high, then the term "slow setback" is used. The latter represents the least urgent safety action. As with reverse, setback ceases as soon as the condition responsible for it has been corrected.

6.110. Since safety signals may come from a variety of instruments, such as ionization chambers, period meters, flow meters, thermocouples, manual switches, etc., located at various parts of the reactor, the system must be designed so as to permit any one signal to cause the appropriate safety action irrespective of the other signals. For situations that do not require instantaneous action, this is relatively simple. All that is necessary is to connect each indicating device to a relay of which the contactor is normally closed. The contactors are then arranged in series, so that a current passes through the circuit as long as no signal requiring safety action is being received. Any one such signal, however, will cause the associated relay to open its contactor and thus interrupt the current. The necessary safety action will then follow.

6.111. Where very rapid action is necessary, as in conditions calling for fast scram, a type of continuous computer called the "sigma" or "auctioneer" circuit has been used. The various devices, e.g., several safety ionization chambers and period meters, from which urgent safety signals might originate are each connected, through suitable amplifiers, to the grids of triode tubes which act as cathode followers. The cathodes of all these tubes are connected together in what is called a "sigma" bus (Fig. 6.10). When the grid of any one of

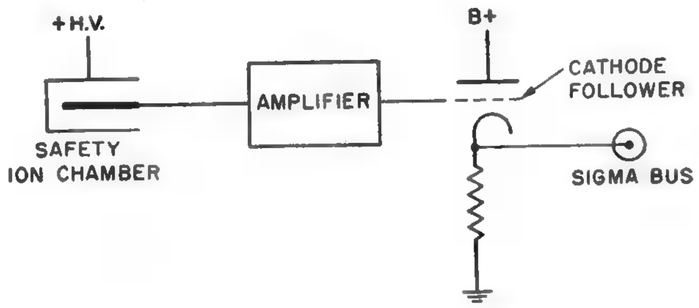


FIG. 6.10. Sigma safety circuit

the triodes receives a signal indicating that safety action is necessary, the cathode goes positive; because of their interconnection through the sigma bus, the cathodes of all the triodes then go positive. The receipt of a safety signal from any one of the amplifiers connected to the sigma bus therefore results in all the others being cut off. The sigma bus thus tends to go along with the "highest bidder" of the various amplifiers; it is for this reason that the designation "auctioneer" circuit has been used.

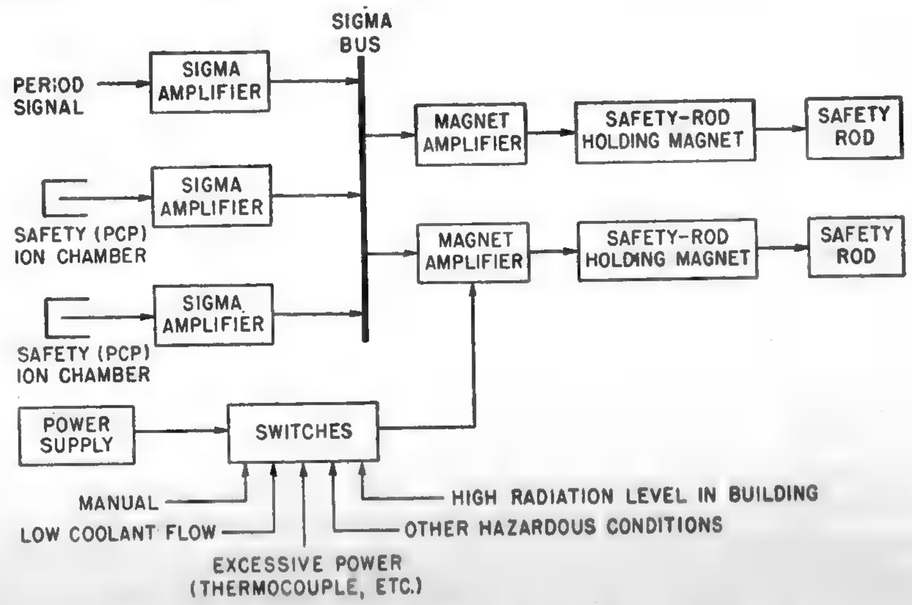


FIG. 6.11. Block diagram of reactor safety system

6.112. The manner in which the sigma circuit is employed in connection with a safety system using electromagnetic clutches and magnet amplifiers, as described above, is indicated in Fig. 6.11. Signals of neutron flux from a number of parallel-plate (PCP) ionization (safety) chambers and of period from a period meter associated with a compensated chamber are fed, through sigma amplifiers, to the sigma bus. The arrival of an urgent signal from one amplifier causes the output current in each of the magnet amplifiers to decrease rapidly. As a result all the safety rods are dropped simultaneously, so that a fast scram occurs.

TABLE 6.4. AUTOMATIC SAFETY ACTIONS IN REACTOR

Condition	Fast Scram	Slow Scram	Reverse	Fast Setback	Slow Setback
Neutron level 150% of full power*	x				
Neutron level 120% of full power*			x		
Neutron level 110% of full power*					x
Period 1 sec	x				
Period 5 sec			x		
Average power 150% of full power†		x			
Average power 120% of full power†				x	
Average power 110% of full power†					x
Coolant flow 80% of normal		x			
Loss of line voltage		x			
Manual		x	x	x	x

* Neutron level as indicated by safety ionization chambers.
† Average power as determined from boron thermocouples or from rate of flow of coolant and temperature rise.

6.113. Some indication of the circumstances under which various safety actions would occur is provided by the summary in Table 6.4. It will be noted that "fast scram" is initiated by only two signals, namely, a neutron level in the reactor 50 per cent above full power and the very short period of 1 sec. The less hazardous period of 5 sec results only in reverse. By manual action, through relays, the operator can cause any safety action other than fast scram.

6.114. Mention should be made of the fact that the description of the safety system given above is for one that would be used for a reactor of fairly high power and advanced design, for which continuity of operation is an important consideration. For reactors of low power, such as are used for experimental purposes, or those having only a small excess reactivity, the only safety action is scram, i.e., complete shut-down of the reactor. The emergencies in such reactors are usually not serious, and since the reactor can generally be restarted relatively easily, the simplest procedure is to shut it down, if necessary, while adjustments are made. In the event of a minor emergency the operator may be warned by an annunciator, e.g., a light signal, so that corrective action may be taken without shutting the reactor down completely.

6.115. In addition to the various abnormal conditions referred to in Table 6.4 that initiate automatic safety action, there are a number of other circumstances which require correction and which should, in any event, be called to the attention of the operator. Among such circumstances are the following: high (local) fuel temperature, excessive radioactivity of the coolant, high level of radiation in the reactor building, ineffective cooling of moderator or shield, and failure of external equipment, e.g., pumps, heat exchanger, etc.

Start-up Accident*

6.116. An essential consideration in the design of a safety system is the matter of the response time. In a reactor designed for high neutron flux and having considerable excess reactivity, the reactor period can become quite short during start-up. If, as a result of operator or mechanical failure, this is allowed to continue through the prompt-critical stage, the power will very rapidly exceed the normal operating level. It is necessary, therefore, in order to obviate the hazard of a start-up accident, that the safety system should respond promptly to period and neutron flux signals. An approximate indication of the permissible time delay may be obtained from the inequality

$$T > \sqrt{\frac{l}{2r \ln (\phi_2/\phi_1)}}, \tag{6.116.1}$$

where T is the "instantaneous period" for the rise of neutron flux, l is the mean lifetime of thermal neutrons in the reactor, r is the rate of change of the multiplication factor (cf. § 4.51), and ϕ_1 and ϕ_2 are the values of the neutron flux at start-up, i.e., of the source, and when safety action is initiated, respectively. Although the theoretical basis of this expression is somewhat uncertain, it has nevertheless been found to be a useful preliminary guide in the design of safety systems for periods less than 50 millisec and for values of r of about 10^{-3} per sec.

6.117. For a reactor using enriched fuel, the neutron lifetime, l, may be of the order of 10^{-4} sec (see § 3.164) and r during start-up may be taken as 10^{-3} per sec, i.e., the multiplication (or reactivity) is increasing at the rate of 0.1 per cent

* H. W. Newson, unpublished; see also, H. Hurwitz, Jr., *Nucleonics*, **12**, No. 3, 57 (1954).

per sec. The ratio ϕ_2/ϕ_1, which is roughly the ratio of neutron level at full power to the source value, would be 10^{12} or more. It follows, therefore, from equation (6.116.1), that under these conditions the minimum period would be about 40 millisec. This means that the neutron flux would be increasing by a factor of 2.7 every 40 millisec when full power is reached and little time is left to check the rise before the reactor is damaged.

6.118. If the response time of the safety system were equal to the minimum reactor period, e.g., 40 millisec in the case under consideration, the power would still rise to 2.7 times the "safe" level before the rod begins to drop. The rate of growth of the flux would then begin to decrease, although the flux itself would continue to increase for some time (Fig. 6.12), since complete insertion of the safety rods cannot be instantaneous. The maximum power attained might well be four or more times as great as the normal operating level, but since the high level would last for a very short time (60 to 70 millisec), the damage to the reactor would probably be negligible. It would seem advisable, therefore, that the response time of the safety system should be of the same order as, or less than, the minimum period of which the reactor is capable. Since the latter depends on the rate of increase of reactivity during start-up, i.e., on the value of r in equation (6.116.1), this quantity should be strictly limited so as to be compatible with the response time. However, it may be noted that a ten-fold decrease in r, i.e., in the rate of withdrawal of the shim rods during start-up, will increase the minimum period by a factor of only $\sqrt{10}$, or about three-fold.

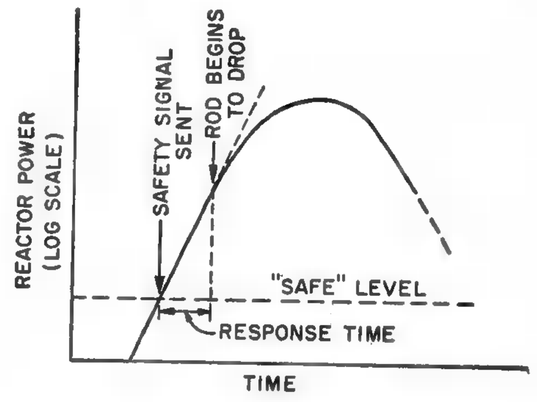


Fig. 6.12. Response of reactor power to safety signal

THE AUTOMATIC LOOP

Functions of Automatic Control System

6.119. Some general aspects of the automatic loop have been already discussed, but a more detailed description of the possible functions and design of the automatic loop will be given here. For reactors of relatively low flux and small excess reactivity, there is no problem of xenon accumulation after shut-down and there is no special reason for a rapid start-up. The danger of a short reactor period is small and the transient fluctuations when operating at full power are not very significant. Operator control of such a reactor, e.g., the Oak Ridge X-10 reactor, is completely adequate.

6.120. For high-flux systems, however, and especially those intended for

mobile power purposes, rapid start-up is essential, and short periods are possible. Further, transients may be large and rapid, because of the excess reactivity built into the reactor. In these circumstances, the maximum degree of automatic control is desirable. Strictly speaking, automatic control includes inherent control due to the negative temperature coefficient of the reactor as well as the use of external devices, such as a servomechanism. The latter aspect of automatic control will be considered first and the effect of the temperature coefficient of reactivity will be examined later.

6.121. Automatic control requires accurate and rapid instrument response, and so it is feasible only in the period and power ranges. In principle, the automatic system would be somewhat as follows. In the period range, the information from a period meter is matched against a particular demand representing the required constant period, and the resulting error signal is fed into the amplifier of a servosystem capable of causing movement of a regulating rod. Any difference between the actual period and the required value will then result in such movement of the rod as will tend to diminish the difference. When the two periods agree, motion of the rod will cease. Automatic period control of this kind could continue right through the power range until the normal power level of the reactor is approached. Control would then be taken over by the neutron level: the signal from an ionization chamber is compared with a reference voltage, representing the required power demand, and the error signal again applied to a servosystem. In this way, the power level of the reactor would be kept essentially constant at a predetermined value.

Methods of Automatic Control

6.122. The application of the foregoing principles involves a number of problems. One of these arises from the fact that period control and level control cannot be independent. Although a constant period is desirable when the reactor is being brought up to power, the period must obviously become infinite when the desired power level is reached, so that the power does not continue to increase. One way in which this could be achieved is illustrated in Fig. 6.13. A single demand voltage, say 10 volts, is used, corresponding to either a specified period, e.g., 20 sec, or to an appreciable fraction, e.g., 10 per cent, of full power; a smaller demand voltage would mean a larger reactor period or a lower power level. The demand voltage is matched against the information from a period detector and a level detector in the reactor, so that any error signal causes a movement of the control rods.*

6.123. As long as the neutron level is well below full power, the information from the level detector will be negligible, and the period detector will be in control of the servomechanism. After the initial stage, when the period is relatively long, the neutron flux will then increase with the constant specified period of 20 sec. When the operating level is approached, some information

* M. A. Schultz, Report AECD-3168.

begins to come from the level indicator. It is this signal, in addition to that coming from the period detector, which now balances the 10-volt demand voltage. The output of the period detector is consequently decreased auto-

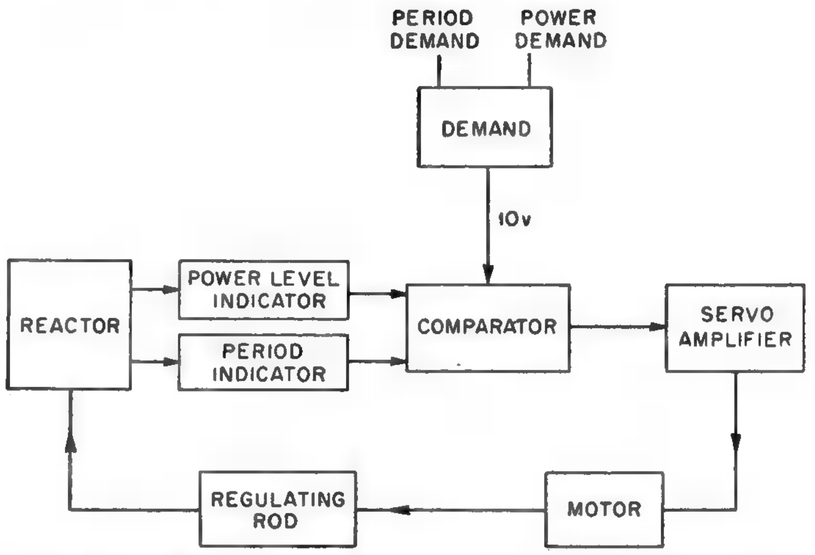


Fig. 6.13. Block diagram of automatic reactor control system

matically, and the reactor operates on a longer period. As the power level increases, the servosystem receives less and less information from the period meter, and more and more from the level indicator. The period thus increases to infinity, and the power levels off to a constant value. The change in power output with time, from start-up to full power level, is indicated qualitatively in Fig. 6.14. It is seen that the lengthening of the reactor period as the desired power level is approached means a gradual leveling off of the power with no tendency to "overshoot."

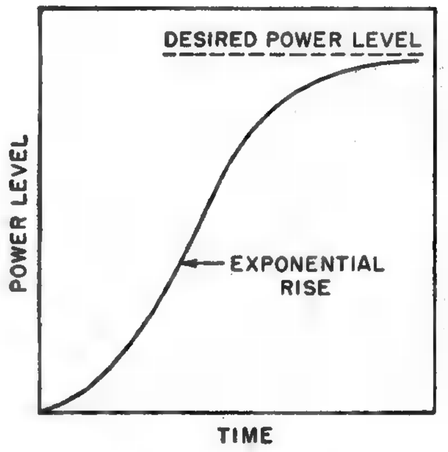


Fig. 6.14. Change in reactor power level with time

6.124. The example considered above represents one of the many different automatic control systems which are possible. One of the difficulties of putting such systems into practice arises from the fact that the time constants of the period and neutron-level circuits are different. As a general rule, the time constant associated with the period information is greater than that for the level signal. A servosystem designed to maintain the reactor level constant should respond rapidly to power level information. But such a system might be useless to control the reactor

period, because of the longer time constant involved in the period indicator circuit. If the time constants differ appreciably, the servosystem may be designed so as to have different response times according to the type of information, period or level, which is in control. Alternatively, two separate servosystems may have to be employed.

6.125. The difficulty may be overcome by the use of a continuous computer system, represented in Fig. 6.15, in which a single servosystem operates entirely from neutron level signals. The motor-operated rheostat is essentially a

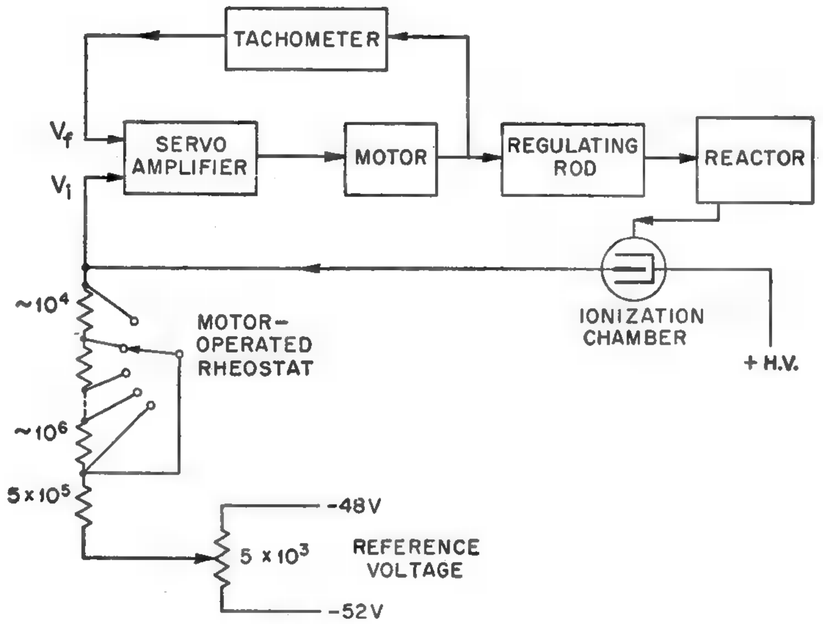


FIG. 6.15. Block diagram of servomechanism for automatic control of period and power level

potentiometer with an exponential winding. This winding is not continuous but consists of 250 steps, the resistance of each step being proportional to the total resistance at that step. The contact, driven by a constant speed motor, acts as the control (or demand) set point for the servomechanism. Thus as the contact moves from bottom to top of the rheostat the resistance decreases exponentially and increased current is required from the ion chamber in order to develop a voltage across the rheostat sufficient to cancel the (constant) negative reference voltage.

6.126. The error voltage, V_i, equal to the sum of the reference voltage, on the one hand, and the voltage developed across the rheostat by the current from the ionization chamber, which is proportional to the neutron flux in the reactor, on the other hand, is applied to the servoamplifier. As a result, a rod-driving motor is energized and a regulating rod is moved, in or out of the reactor, at a speed proportional to the error voltage. When the current from the ion chamber is less than that required to develop a voltage across the rheostat equal in magnitude to the negative reference voltage, the sign of the error voltage is the same as that of the reference voltage. This causes rod withdrawal and the neutron flux level increases. If, however, the voltage across the rheostat is larger than the reference voltage, the sign of the error voltage is reversed; the regulating rod is then inserted and the neutron flux decreases. The servomechanism thus operates in such a manner as to keep the ion current, and hence the reactor power level, up to the demand set by the rheostat position.

6.127. As the contact moves up the exponential rheostat the current demanded from the ion chamber to attain zero error voltage increases exponentially, as stated above, and so the servomechanism adjusts the position of the regulating rod in such a manner as to increase the neutron flux exponentially. In other words, in view of equation (4.9.1), the neutron level of the reactor will be increasing on a constant period, the value of which is determined by the speed of the motor moving the contact over the rheostat.

6.128. It will be seen from Fig. 6.15 that a feedback voltage V_f is also applied to the servoamplifier. This voltage is derived from a tachometer attached to the regulating rod drive, and so it varies in proportion to the speed of the regulating rod motion. The purpose of the feedback voltage is to damp the movement of the rod and thus prevent overshoot. Actually when the reactor power level is rising on a constant period the reactivity will be constant and the regulating rod will remain essentially stationary.

6.129. When the moving contact has reached a position on the rheostat corresponding to the desired power level, the motion of the contact can be stopped and fine adjustment of the demand level is then made by the 5000-ohm (vernier) potentiometer shown at the bottom of Fig. 6.15. This potentiometer, which determines the reference voltage, is located on the reactor control console and is operated in connection with a meter indicating the average reactor power. As long as the reactor is at the desired power level the voltage developed by the ion chamber will balance the reference voltage and there is no error voltage. Any deviation in the reactor power, due to transient changes, immediately causes an error signal to be applied to the servoamplifier and the regulating rod is automatically moved to correct the change in power level.

6.130. Attention should be called to the fact that, although the automatic system described above, based on neutron flux level signal, is relatively simple, it has the disadvantage that its use as a satisfactory period control is restricted to the power range of operation, where level signals are reliable. If, in addition, control in the period range, from 10^{-5} to 10^{-2} of full power, is required, as it might be for a mobile power system, it appears that period information must be included. In reactors having only a small excess reactivity, such as the Water Boiler, there is no real need for period control. The automatic system is then used for the sole purpose of maintaining the level constant once the reactor has been brought up to operating power by manual control.

Design of the Servosystem

6.131. Although a detailed discussion of the design of the servosystem lies outside the scope of this book,* there are certain aspects of the subject, based on the time behavior of reactors, which merit consideration here. It will be seen from Fig. 6.1 that it is possible to go around the automatic loop either by passing directly from reactor to instruments or by going from the reactor through the external load to the instruments. In the first place the load will be ignored, i.e., it will be assumed constant, but its effect will be considered later.

6.132. Let n_0 be the neutron density to which the reactor is to be controlled by the automatic system, and suppose the corresponding value of the effective multiplication factor is k_0. Then, if n and k are the actual values of the neutron density and the multiplication factor at any time, the deviations are represented, as in § 4.63, by

$$n' = n - n_0 \quad \text{and} \quad k' = k - k_0.$$

In this case n' is the quantity to be controlled, since it is a measure of the deviation from the desired condition, and k' is the parameter whereby the control is to be effected, by the movement of a regulating rod.

6.133. In order to determine what function of n' might be used as a basis for the design of the servosystem, an examination is made of the differential equation in n with respect to time. By combining equations (4.64.1) and (4.64.2), which apply when the changes in n and k are small, it is possible to eliminate the C_i''s; assuming, for simplicity, the approximate case of one (average) group of delayed neutrons, and writing the operator p to represent d/dt, the result is

$$\left[\frac{l}{\lambda}p^2 + \left\{\frac{1 + l\lambda - (1-\beta)k_0}{\lambda}\right\}p + 1 - k_0\right]n' = n_0\left(\frac{1-\beta}{\lambda}p + 1\right)k', \tag{6.133.1}$$

where λ is the weighted mean life of the delayed neutrons, i.e., 0.08 sec^{-1} (§ 4.22). In the present circumstances, the neutron density n_0 is that for the stationary state of the reactor, so that k_0 is unity; consequently equation (6.133.1) reduces to

$$\left(\frac{l}{\lambda}p^2 + \frac{\beta + l\lambda}{\lambda}p\right)n' = n_0\left(\frac{1-\beta}{\lambda}p + 1\right)k'. \tag{6.133.2}$$

6.134. In considering this differential equation, it is seen that n_0, the demand value of the neutron density, appears on the right side. In order to remove this term it is necessary either that the control parameter vary inversely with n_0 or that dynamic control, which varies its quality with n_0, be employed. In general, the former alternative is chosen, so that the control parameter is made a function of n'/n_0. A simple error-actuated (or proportional) control, such as

* See references to § 4.63; also, J. H. Bowen, *Proc. Inst. Elec. Eng.* (*Lond.*), **100**, Part I, 102 (1953); J. M. Harrer and J. A. Deshong, Jr., *Nucleonics*, **12**, No. 1, 44 (1954).

is frequently used, would then make k' proportional to n'/n_0, i.e.,

$$k' = -a\left(\frac{n'}{n_0}\right). \tag{6.134.1}$$

Since, over a considerable range of the regulation rod motion, the change in the reactivity (or multiplication) is proportional to the distance the rod is moved (§ 6.52), equation (6.134.1) is the basis of what is called a "position" servomechanism. The amount of change in position of the rod is proportional to the error function.

6.135. Upon substitution of equation (6.134.1) into (6.133.2), it is found that

$$[lp^2 + \{\beta + l\lambda + a(1-\beta)\}p + a\lambda]n' = 0. \tag{6.135.1}$$

The controlled reactor is thus seen to respond as a second order system with the following characteristics:*

$$\text{Natural angular frequency} = \sqrt{\frac{a\lambda}{l}}$$

$$\text{Damping ratio} = \frac{\beta + l\lambda + a(1-\beta)}{2\sqrt{al\lambda}}.$$

6.136. If a were taken to be 1 and the mean lifetime, l, of the thermal neutrons in the reactor is 10^{-3} sec, then, since λ is 0.08 sec^{-1} and β is 0.0075, the natural frequency is found to be about 9 radians/sec, i.e., 1.5 cps, and the damping ratio is 55. With such a large damping ratio the response of the system would be extremely sluggish. By decreasing the proportionality factor a the damping ratio passes through a minimum, but the lowest value is 9.7, with a natural frequency of 0.78 radian/sec, i.e., 0.13 cps. The system thus behaves essentially as one of the first order, and from equation (6.135.1) it follows that

$$\text{First order time constant} = \frac{1-\beta}{\lambda} + \frac{\beta + l\lambda}{a\lambda},$$

the minimum value of which, for large a, is $(1-\beta)/\lambda$, i.e., 12.4 sec.

6.137. It is evident, therefore, that the simple proportional control, represented by equation (6.134.1), must inevitably lead to sluggish performance and so is not suited to automatic reactor control. There is also another reason why a position servomechanism would not be too desirable for this purpose. Because of fuel depletion, accumulation of fission product poisons, temperature changes, etc., the excess reactivity of a reactor does not remain constant during operation. Hence the position of the regulating rod, corresponding to a specified power level, must be continuously adjusted to compensate for this variation.

6.138. Another possibility is to make the time rate of change of the control parameter (rather than the parameter itself) vary as the control function n'/n_0, so that the control formula would be

* J. D. Trimmer, "Response of Physical Systems," John Wiley & Sons, Inc., 1950.

$$\frac{dk'}{dt} = -a\left(\frac{n'}{n_0}\right). \qquad (6.138.1)$$

This is sometimes referred to as integrating control, since the control parameter k' is proportional to the integral of $(n'/n_0)\,dt$. Upon substituting equation (6.138.1) into (6.133.2), and recalling that p represents the operator d/dt, the result is

$$[lp^3 + (\beta + l\lambda)p^2 + a(1 - \beta)p + a\lambda]n' = 0. \qquad (6.138.2)$$

This third order equation can be solved graphically, by separating it into two parts, one involving p^3 and p^2 and the other containing p only. The solutions for two values of a are given below.

	$a = 0.001$	$a = 0.05$	
Time constant (first order part)	0.134	12.5	sec
Natural frequency (second order part)	0.104	7	radians/sec
Damping ratio (second order part)	0.635	0.535	

6.139. Although neither of these two combinations of time constant, natural frequency, and damping ratio would give ideal response, the integrating control, represented by equation (6.138.1), is considerably better than the simple proportional control of equation (6.134.1). Incidentally, since the former is a velocity control rather than a position control, it will also take care of the point, mentioned above, that the position of the control rod must change even during operation at constant power.

6.140. Attention may be called to the fact that the servosystem for reactor and period control depicted in Fig. 6.15 is based on the integrating control formula. The circuit is such that the error voltage V_i is proportional to n'/n_0. Since the regulating rod is moved at a rate proportional to the error voltage, and the rate of motion, over a considerable range, varies directly as the change in the multiplication factor, it follows that the time rate of change of the latter is proportional to n'/n_0, in accordance with equation (6.138.1).

6.141. The purpose of the foregoing treatment is to indicate the approach that may be used in the analysis of an automatic control formula. Although the integrating control is used in certain reactors, other types of control are possible. If the system is to function over an appreciable range of power, i.e., of n_0, then it is desirable that the control parameter shall be inversely related to n_0. However, if the automatic control is required to operate at a single power level only, so that n_0 is constant, the control parameter can be merely a suitable function of n', the difference between the demand power and the actual value.*

* For general discussions of automatic control systems for reactors, see M. A. Schultz, *op. cit.*; J. M. Harrer and J. A. Deshong, Jr., *loc. cit.*; J. H. Bowen, *loc. cit.*; R. V. Moore, *Proc. Inst. Elec. Eng.* (*Lond.*), **100**, Part I, 90 (1953); also, E. Fermi, *Amer. J. Phys.*, **20**, 537 (1952), see page 557.

6.142. Because of the nonlinearity of the kinetic equations the device was used above of considering small variations from an equilibrium condition. This approach is useful as a preliminary guide, but it does not provide a complete basis for the design of a servosystem for automatic control. To proceed further it is necessary, short of building and experimenting on the reactor itself, to make use of electrical reactor-simulators, as described in Chapter IV, for testing the operation of the servomechanism. Although a perfect simulator does not exist, the use of such an instrument represents an important step between a mathematical analysis of the type given above and actual trial of the system on the reactor itself.

Self-regulation by Temperature Change

6.143. It was seen in § 4.74, *et seq.*, that temperature changes affect the reactivity of a reactor. Consequently alteration in temperature might possibly contribute to the control of the system. In general, essentially all reactors have negative temperature coefficients, so that if the temperature rises, e.g., because of an interruption in coolant flow, the reactivity will decrease.* There will be a corresponding decrease in the power level which will tend to compensate for the increase of temperature. The task of control design is thus considerably simplified if the reactor has this self-regulating feature. It is particularly advantageous if an important contribution is made by the coefficient of reactivity arising from the fuel itself, since this responds promptly to changes in the temperature.

6.144. Advantage is being taken, both in existing and planned reactors, of the self-regulating effect of temperature. It helps to compensate automatically for power variations due to local transient changes in the reactor as well as to changes in the entry temperature and flow rate of the coolant. Largely because of the high temperature coefficient of thermal expansion, reactors employing water, either ordinary or heavy water, as moderator and coolant have exceptionally large negative temperature coefficients of reactivity (§ 4.91). For the Water Boiler, for example, the value is -3×10^{-4} per °C (-1.7×10^{-4} per °F) and coefficients approximating -10^{-3} per °F are possible for water-moderated and -cooled reactors operating at higher temperatures. Such reactors consequently have the self-regulating property to an exceptional degree.

6.145. It may be remarked that the temperature effect can compensate for the steady decrease in reactivity arising from burn-up of fuel and the accumulation of fission product poisons only by a corresponding decrease in operating temperature. To compensate for these changes and maintain constant temperature the movement of a control rod is required, unless fuel can be added during operation.

* Reactors in which water is the coolant but is relatively unimportant as the moderator are exceptional in this respect (see § 4.92).

The External Load and Automatic Control*

6.146. The question of the effect of the external load on the automatic control system is important when significant and variable amounts of power are being drawn from the reactor. It will be supposed, as is the common situation for power reactors in existence or under construction or development,† that the coolant (or fluid fuel) is circulated through a heat exchanger in which steam is produced for the operation of a turbine, as indicated in Fig. 6.16.

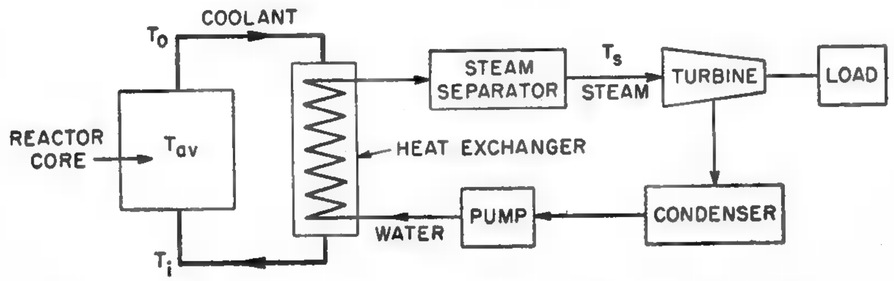


Fig. 6.16. Relationship between load and reactor control

6.147. If the rate of flow of coolant (or fluid fuel) in the primary circuit is constant, then the power output of the system, P, may be represented, to a good approximation, by

$$P = K(T_{av} - T_s),$$

where K is a constant; T_{av} is the average temperature of the coolant in the reactor and T_s is the steam temperature at the throttle. Although, in principle, the program of the system may be such that variations in both T_{av} and T_s are possible, two extreme cases will be considered first; these are (1) T_{av} constant, and (2) T_s constant.

Constant Average Coolant Temperature

6.148. In this case the power output is varied by changing the steam pressure at the throttle and the steam temperature, T_s, changes correspondingly. An increase in power demand, for example, will result in a decrease in T_i, the inlet temperature of the coolant to the reactor. Since this will cause a lowering of the reactor temperature the reactivity increases, because of the negative temperature coefficient, and the power output will rise to keep pace with the demand. The outlet temperature, T_o, will increase so as to maintain T_{av} constant, as postulated. Thus throttle changes are automatically communicated to the reactor as changes in reactivity which cause it to vary its power output automatically in a stable manner to meet the demand. No control rod motion is

* M. A. Schultz, *Proc. Conf. on Nuc. Eng.*, Berkeley, 1953, D-1.
† Boiling-water reactors are exceptions (§ 8.238).

required except, as stated in § 6.145, such as may be needed to allow for fuel burn-up and poisons.

6.149. The program of constant average temperature of coolant is thus the natural one for operation of the reactor, but it has disadvantages as far as the steam system is concerned. The large variations of steam pressure which occur in the secondary system necessitate special turbine design and automatic throttling valves are required to feed a constant-frequency turbine generator. Further, steam pipes of large size are necessary and difficulties arise in connection with the boiler-feed pump.

Constant Steam Temperature (and Pressure)

6.150. The program of constant steam temperature (and pressure) is ideal for the secondary system, since it permits optimum design of the steam plant. Among the advantages are the following: smaller steam pipes can be used, the turbines are nearly standard, and neither special boiler-feed pumps nor automatic throttling valves are needed.

6.151. The disadvantages of the constant steam pressure system are felt, however, by the reactor. Any change in the power demand at the throttle now means a change in T_{av}, since T_s is constant. The self-regulating tendency for the reactor to maintain a constant average coolant temperature must then be compensated by a relatively large amount of rod motion. When the power demand is changed the action of the automatic control system is antagonistic to the effect of the negative temperature coefficient of the reactor.

6.152. It might appear that a good engineering compromise could be achieved by allowing the steam pressure (and temperature) to decrease somewhat with the load and, at the same time, permit the average temperature of the coolant to rise somewhat. However, a better solution has been proposed; the system is designed so that for steady-state operation it behaves as a constant pressure steam plant, while for transient changes it operates as constant average coolant temperature system. This is achieved by design of the automatic control system with such characteristics that there is a relatively slow movement of the regulating rod in response to a change in the neutron flux signal.

6.153. If the throttle is changed slowly, at constant steam pressure, i.e., in essentially steady-state operation, the control rod moves slowly to keep the power level of the reactor equal to the demand. On the other hand, if the throttle is opened or closed suddenly, there will be a rapid change in the inlet temperature of the coolant. But since the regulating rod moves slowly the negative temperature coefficient of the reactor supplies the change in reactivity required to match the transient change in the load. For sudden throttle changes of short duration the reactor thus behaves as if it were operating without control rods and is stabilized by the self-regulating effect of the reactivity temperature coefficient. If the system now continues to operate with the new power load, at constant steam pressure, control of the reactor passes to the regulating rods.

6.154. Some preliminary information concerning the design of a control system to satisfy the foregoing requirements can be obtained by analytical methods. The final design is best accomplished, however, by means of a reactor simulator, as stated above.

REACTOR OPERATION

Initial Start-up

6.155. Normal operation of a reactor may be divided broadly into three parts: start-up, power operation, and shut-down. It is during start-up, in particular, that the problem of control requires careful attention. Actually, three different types of start-up may be distinguished. There is, first, what may be called "initial start-up," frequently referred to as start-up from "cold," which applies to the starting of a new reactor, one which has been refueled, or one which has been shut down for several days. Second, there is start-up within a short time after accidental or deliberate shut-down; this is sometimes called "start-up after scram." Finally, it may be necessary to start up the reactor after a reduction in power, e.g., to about 0.1 or 1 per cent of full operating power; this has been designated as "power range start-up."

6.156. All start-up operations have the same objective, namely, bringing the reactor to critical and then increasing the neutron flux (and power output) in a carefully controlled manner until the desired level is reached. Initial start-up requires special attention because the neutron level may be so low that measurements with the usual instruments are not very reliable. In any event, it is of the utmost importance that no control rod be moved until the neutron flux is large enough to be detected, on a counting-rate meter at least. This detectable flux may arise from spontaneous fission or it may result from the inclusion of a special neutron source in the reactor, as will be explained below (see also § 5.112).

6.157. Even in these circumstances, there is a possibility that the reactor may go through prompt critical on a short period; this situation can be avoided by careful regulation of the rate of increase of reactivity, i.e., of the rate of withdrawal of the control rods, during initial start-up. An examination of Fig. 6.17 will prove instructive in this connection. It gives the reactor period * as a function of the time after start-up for various rates of increase of reactivity. The initial effective multiplication factor was taken to be 0.9 in each case, and the neutron lifetime in the reactor was 10^{-4} sec. It is seen that, when the rate of change of reactivity is 0.002 per sec, the reactor becomes critical after 50 sec and the period is then only about 1.5 sec. However, by increasing the reactivity more slowly, e.g., 0.0005 per sec, the period when the reactor becomes critical is some 7 sec. Whereas the former period is dangerously short, the latter is relatively safe.

* The term "period," as used here, is the reciprocal of $d \ln n/dt$ at a particular instant; it is not a true (stable) reactor period.

6.158. For the large, natural uranium-graphite reactors, such as the Brookhaven and Oak Ridge X-10 reactors, the chances of going into a short period during start-up are not great. There are several reasons for this situation. In the first place, these reactors have relatively small excess reactivity. Further, the reactivity equivalent of the control rods is not large, and so with any reasonable rate of rod withdrawal the rate of increase in the multiplication factor of the reactor will be small. Finally, the neutron lifetime in a natural uranium reactor is fairly long, and so a given reactivity if greater than 0.005 will correspond to a considerably longer period than would be the case if the lifetime were shorter (see § 4.38).

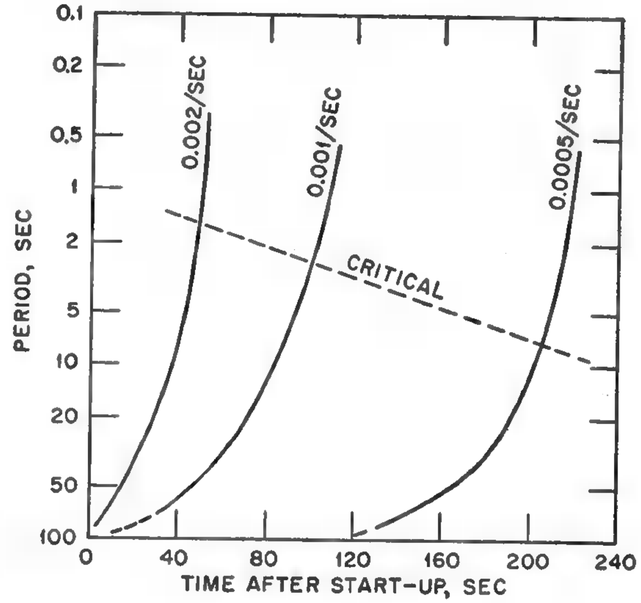


Fig. 6.17. Reactor period as function of time for various rates of increase of reactivity

6.159. In natural-uranium reactors, too, the neutron source from the spontaneous fission of uranium-238 is sufficient to permit safe start-up without an additional source of neutrons. Spontaneous fission yields about 15 neutrons/sec per kilogram of natural uranium, and this produces a source strength of about 5×10^5 neutrons/sec in the core of the X-10 reactor. Since various instruments are available which will respond to this source strength, the neutron level can be readily followed during start-up.

6.160. A reactor of high power, employing enriched fuel, such as that referred to in Table 6.1, will usually have a large excess reactivity, possibly 0.25 or more. The too rapid withdrawal of the control rods would then result in a high rate of increase in the multiplication factor of the reactor. This combined with the small neutron lifetime could result in an extremely short and highly dangerous period. Very special precautions must therefore be taken in the start-up of such a reactor.

6.161. Because of the small total mass of fuel in an enriched reactor, as compared with one employing natural uranium as fuel, and the small amount of uranium-238 present, spontaneous fission alone will not produce neutrons at a sufficient rate for their detection to be possible. It is consequently necessary that an additional neutron source, e.g., one consisting of polonium and beryllium (§ 2.72), be built into the reactor, for initial start-up at least. Sources such as this yielding 10^6 to 10^7 neutrons/sec are readily available, and so an adequate indication on a fission counter or a proportional counter containing boron is possible before the control rods are moved. It is thus feasible to follow the neutron level by means of suitable instruments over the entire range, from start-up to the operating power level. The possibility of a very rapid rise in the neutron flux, i.e., a very short period, without the operator being aware of the situation, is thus very greatly decreased.

6.162. Because instrumentation at low neutron levels, at the lower end of the counter range, is very unreliable, it is not advisable to withdraw the control rods continuously. Intermittent withdrawal, e.g., 0.5 sec on and 4.5 sec off, as is employed in the MTR, however, provides an important element of safety. As long as the reactor is subcritical, the neutron flux will increase during the "on" period, due to subcritical multiplication, but will tend to become constant during the "off" period (§ 4.66, *et seq.*). When the reactor passes through the critical state, the flux continues to increase even during the "off" period. In this way, the operator will become aware that the system has gone critical, and he can then take precautions to prevent the reactor "running away."

6.163. Up to this point, the control has been strictly manual *; because of the unreliability of instruments at low neutron levels, there appears to be little prospect at present of making initial start-up purely automatic. When the flux has increased to the point that fairly satisfactory period information is available, i.e., in the period range, it is possible to substitute continuous rod withdrawal for the intermittent procedure. The period meter will then serve as a guide to control, which can be either manual or automatic, until the power range is attained. As explained earlier, neutron level information is used to bring the reactor up to its normal power.

Start-up After Scram

6.164. Start-up after a brief shut-down is a much less hazardous process than initial start-up. If the delay has not been too great, the neutron level will probably be high enough to give good period information. There are several factors contributing to this situation. One is the limiting effect of the delayed neutrons, so that even 10 min after shut-down the neutron flux will have decreased by a ratio of only about 10^5 and the reactor will still be in the period range. Another factor is the formation of neutrons by the action of the fission-

* The term "manual," as contrasted to "automatic," implies that the human operator is in charge of control-rod adjustments.

product gamma radiation upon various reactor materials. This will be particularly important when heavy water or beryllium is employed as moderator or reflector (§ 2.74).

6.165. For some time after shut-down the subcritical reactor will thus behave as if it contained a large primary source. When the reactor is started up again, this large source and the subcritical multiplication will give a neutron flux for which reliable instrumentation will be possible even prior to withdrawal of the control rods. In the MTR, the intermittent start-up can be eliminated, and continuous rod withdrawal permitted, provided the neutron flux is at least 10^{-5} of the normal operating level, since period control is then possible. The beryllium reflector used in this reactor facilitates the attainment of this condition.

6.166. Because of the growth of the xenon concentration after shut-down in a high-flux thermal reactor, it may be desirable to start up again as soon as possible. If this is done within a short time, the start-up would be carried out as described above. However, if the delay is long enough to permit the xenon concentration to grow to such an extent that there is insufficient built-in reactivity available to override the poisoning, then it will be necessary either to replace some of the fuel elements or else to allow the reactor to stand until the xenon has decayed. In this case, as in other instances where the shut-down has been prolonged, the neutron level will have decreased greatly and the start-up procedure will be essentially the same as for initial start-up.

Power Range Start-up

6.167. In certain circumstances the power level of a reactor will be reduced to a small fraction, e.g., 0.1 to 1 per cent, of its full operating power. While this is quite small from the power standpoint, the neutron flux is so high that reliable information is available from both period and neutron level instruments. Start-up under these conditions presents no special problem. It can be performed manually, if desired, or it can be automatically controlled by a servo-system, as already described. All that is necessary is to set a period that will bring the reactor up to its normal power at a reasonably fast rate compatible with safety.

Normal Operation of Reactor

6.168. The function of the control system in normal operation of a reactor requires little further discussion, in view of the references to this matter that have already been made. The essential requirement for a research or experimental reactor is usually that the neutron flux (or power level) remain as constant as possible. The stability of such reactors, e.g., the Oak Ridge X-10 reactor, is often such as to permit satisfactory manual operation, and the addition of an automatic control system is then based on considerations of convenience. For a reactor such as the MTR, on the other hand, the fluctuations introduced by the cooling system and by certain experiments are appreciable

enough to make manual control difficult. The incorporation of a servosystem then aids materially in attaining satisfactory operation.

6.169. For reactors designed as power producers the main requirement during operation is that the reactor follow load fluctuations as they occur. As seen above, the negative temperature coefficient of the reactor will aid materially in this respect, and under proper conditions can make it unnecessary to have an external control system (see § 13.102).

6.170. Among the factors which may cause transient effects in a reactor, that can be readily dealt with by automatic control, are momentary changes in temperature and rate of flow of coolant, due either to external or internal causes. Sudden introduction or withdrawal of neutron absorbers, either accidentally, e.g., in the coolant, or intentionally, e.g., in an experiment, will also affect the reactor equilibrium. In fact, a change in almost any of the conditions in the reactor, such as movement of an instrument, is likely to produce some effect on the power level.

6.171. Apart from the transient phenomena, there are steady changes taking place in a reactor for which continuous, if slow, adjustment is necessary in order to maintain a steady power output. Mention may be made of fuel depletion, accumulation of fission product poisons, and changes in dimensions of structural and other materials due to operation at a high temperature and in a high radiation field over a long period of time.

Shut-down of Reactor

6.172. There are, in general, two ways in which a reactor may be shut down: controlled (or deliberate) and enforced (or scram). If the controlled shut-down is to be of long duration, e.g., for refueling or for maintenance purposes, then the procedure adopted is immaterial. All the control rods may be inserted as rapidly as desired. On the other hand, if the shut-down is expected to be temporary, while an external repair is performed, then the neutron level should be decreased, by gradual insertion of the shim rods, until the power output is the maximum that can be tolerated with safety. This will not only minimize the xenon build-up (§ 4.111), but will also simplify the restart operation. In deciding upon a safe degree of shut-down, it must be remembered that considerable heat will continue to be liberated as a result of radioactive decay of the fission products.

6.173. In order to insure continued operation of the reactor, enforced shut-down (or scram) should be avoided as far as possible. Small fluctuations in power, which may be caused by variation in the coolant, etc., or apparent fluctuations, which may be due to changes in the controlling instruments and their associated circuits, should not be able to produce a scram. Thus, as seen in Table 6.4, the safety system of a reactor can be designed so that the lesser emergencies cause a gradual decrease of power, but not a complete shut-down. Only when the power gets dangerously high, e.g., 50 per cent above the normal

level, or the period becomes dangerously low, e.g., 1 sec, will a fast scram occur automatically.

Symbols Used in Chapter VI

a	constant
d	extrapolation distance
K	constant
k	multiplication factor
Δk	change in multiplication factor
k_0	value of multiplication factor to be controlled
k'	deviation from k_0 ($k' = k - k_0$)
k_{eff}	effective multiplication factor
l	neutron lifetime in reactor
n	neutron density
n_0	value of neutron density to be controlled
n'	deviation from n_0 ($n' = n - n_0$)
P	reactor power output
p	differential operator (d/dt)
R	radius of cylindrical reactor
R_0	radius of cylindrical control rod
r	rate of increase of multiplication factor
T	reactor period
T_{av}	average temperature in reactor
T_i	coolant inlet temperature
T_o	coolant outlet temperature
T_s	steam temperature
t	time
V_f	feedback voltage
V_i	error voltage
β	fraction of fission neutrons delayed
λ	weighted average lifetime of delayed neutrons
ρ	reactivity
ϕ_1	neutron flux at start-up
ϕ_2	neutron flux when safety action is initiated

PROBLEMS

1. The change in reactivity (or multiplication factor) due to insertion of an absorber at any point into a reactor may be taken to be proportional to the square of the thermal-neutron flux at that point. On this basis derive (either analytically or arithmetically) the general shape of the curve in Fig. 6.4, showing the change of reactivity with the extent of insertion of a control rod. The flux may be assumed to have a cosine distribution in the direction of rod motion that is not greatly changed by insertion of the rod.

2. A reactor operating at a thermal flux of 10^{14} neutrons/(cm^2)(sec) has excess reactivity of 0.10 available for overriding xenon poisoning. Suppose the reactor has been

operating for some time and is then completely shut down. During what time interval can the reactor be restarted? If this time has elapsed, how long will it be necessary to wait before start-up is again possible, without refueling? (Use the results of Problem 4.13.)

3. The aqueous fuel solution of a homogeneous reactor has a concentration of 120 grams of uranyl sulfate (UO_2SO_4), enriched to the extent of 90 per cent in uranium-235, in a volume of 1 liter. Estimate the concentration of lithium sulfate (Li_2SO_4) which would be required to compensate for burn-up of the uranium-235, so as to minimize continuous adjustment of the regulating rod during reactor operation. (Use cross section data in the Appendix.)

4. In start-up the multiplication factor of a reactor is increased at the rate of 0.2 per cent per second by the continuous withdrawal of a control rod, which is not checked. When the neutron flux is 5×10^{11} of the original value automatic safety action is initiated. but there is a delay of 50 millisec before the safety rods are inserted. Estimate the extent to which the "safe" level of the reactor is exceeded and the time for which the overload exists. (The neutron lifetime in the reactor is to be taken as 10^{-4} sec.)

5. Calculate the maximum permissible rate of increase of the multiplication factor by control rod withdrawal during start-up, for the preceding problem, if the reactor overload is not to exceed 50 per cent.

6. Derive equation (6.138.2) and verify the results given in § 6.138.

7. Outline a complete control and safety system, including interlocks, such as might be required for a reactor designed to produce large-scale power. (It is to be assumed that the same control rods are used for shim and safety purposes.)

Chapter VII *

PROCESSING OF NUCLEAR REACTOR FUEL

INTRODUCTION

Separations Chemistry

7.1. The applications of chemistry and of chemical engineering in the nuclear reactor field fall into two main categories. The first is concerned with the production of materials required for the construction and operation of the reactor. These include reactor fuels, e.g., uranium and thorium, moderators, coolants, and structural materials. The second involves the treatment of the spent fuel elements, after use in the reactor, for the extraction of new fissionable species, e.g., plutonium, which may have been produced, and for the recovery of the unused fuel. An aspect of this second category is the disposal of the waste solutions containing radioactive fission products.

7.2. In most reactors constructed hitherto, the designs have been based on the most readily available suitable materials, with little consideration of the chemical problems involved in the treatment of the spent fuel. It has been realized that, from the long-range point of view, a lack of coordination is uneconomic, especially as the degree of efficiency achieved in the extraction of the fissionable material may represent the margin between failure and success in breeding (§ 1.165). Consequently a new field of investigation, sometimes referred to as "reactor chemistry," is emerging. It is concerned, among other things, with the development of new fuel-moderator systems, such as homogeneous solutions, slurries, and melts, which have the advantage of either relative simplicity of preparation or of treatment, or both.

7.3. The present chapter is concerned mainly with separations procedures, i.e., with the second of the categories described in § 7.1. In connection with regenerative reactors, either converters or breeders, it is desirable to extract the fissionable material produced during operation and also to recover the unused

* Material contributed by F. R. Bruce, F. L. Culler, W. K. Eister, D. E. Ferguson, J. R. Flanary, S. Glasstone, I. R. Higgins, A. C. Jealous, W. B. Lanham, R. E. Leuze, J. F. Muggs, S. Peterson, J. T. Roberts, R. W. Stoughton, and W. H Sullivan.
Reviewed by W. K. Eister, R. W. Stoughton, and W. H Sullivan.

fuel. The spent fuel elements from the Hanford converter reactors were originally treated only for the extraction of the plutonium; the residual uranium, together with the fission products, was stored in underground tanks. However, the ideal approach would be to separate the plutonium and uranium from each other and from associated fission products. In addition, it would be desirable to recover the uranium from the accumulated residues now in existence.

7.4. In reactors employing fuel consisting of highly enriched uranium-235, there is no appreciable formation of plutonium, and so the spent fuel elements are treated only for the removal of fission products from the uranium. If the enriched reactor has a breeding blanket, then this must be processed to extract the fissionable material, i.e., plutonium-239 or uranium-233, and to recover the remaining fertile material, i.e., uranium-238 or thorium-232, respectively. Since some fission may occur in the blanket, it may be necessary to purify the material from fission products.

Spent Fuel Elements

7.5. The first step in the discussion of separations chemistry is to examine the circumstances which determine the necessity for removing (or discharging) the so-called "spent" fuel elements from the reactor for processing. A limit is set upon the time a fuel element can remain in the reactor by the changes in dimensions and shape that occur with continued operation (see Chapter VIII). There is little doubt that in due course the problems arising from the physical damage caused by radiations will be overcome either by special designs, by the use of suitable alloys, or by employing the fuel in a fluid form. Nevertheless, other conditions will still be operative which will necessitate relatively frequent processing of the reactor fuel.

7.6. If the fuel consists mainly of fissionable material, such as uranium-235, the burn-up (or depletion) of fuel, due both to fission and nonfission capture reactions, and the accumulation of fission product poisons will decrease the reactivity of the reactor to such an extent that the design requirements will no longer be satisfied (see § 6.31, *et seq.*). At this time only a fraction of the fuel will have been destroyed and the spent elements will still contain a large percentage of fissionable material. The fuel elements must therefore be processed in such a manner as to recover essentially all of the remaining fissionable species.

7.7. In a reactor using natural (or slightly enriched) uranium as fuel, so that appreciable amounts of uranium-238 are present, there will inevitably be some formation of plutonium-239. Apart from physical changes in the fuel elements and poisoning due to fission products, the period for which the elements can remain in the reactor will depend upon its primary purpose, namely, power or plutonium production.

7.8. If the main objective is power production, then it may be possible to continue operation for a longer time, since fissionable plutonium-239 replaces the uranium-235 used up, in roughly equal amounts (§ 1.138). On the other hand, if it is desired to recover the maximum amount of plutonium-239, account must be taken of the losses which will occur, due to various neutron absorption reactions, with long exposure times. For a dual-purpose reactor the rate of discharge of fuel elements for processing will be determined by over-all economic considerations.

7.9. In the production of uranium-233 from thorium-232 (§ 1.61) the limiting factor may be either the destruction of uranium-233 by neutron absorption or the accumulation of uranium-232. The latter can be formed in a number of different ways, as can be seen from Fig. 7.1 (below), and since the isotopes of uranium have essentially identical chemical properties the uranium-232 is extracted with the uranium-233. The presence of uranium-232 is undesirable because it is an alpha-particle emitter of fairly short half life (70 yr), and its decay product, thorium-228, is a member of the thorium series of radioelements. Upon standing, therefore, the radioactivity of the material increases, because of the formation of alpha-, beta-, and gamma-active decay products, thus making it more difficult to handle.

Heavy Isotope Build-up

7.10. In a reactor containing (or producing) plutonium-239 some neutron capture will occur, since the cross section is relatively high (Table 2.10); this will result in the formation of plutonium-240. The latter can, in turn, also capture neutrons to yield plutonium-241. This isotope undergoes beta decay, as well as neutron capture, so that other species are formed. An indication of

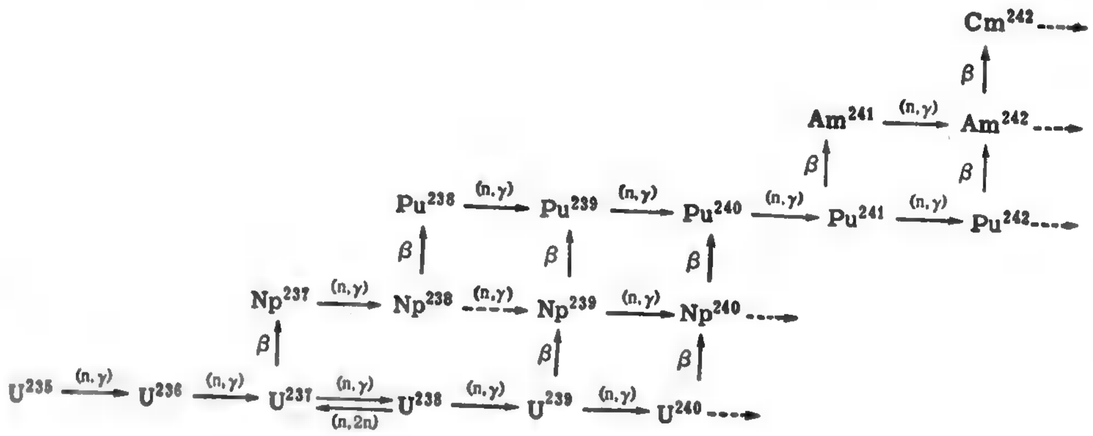


Fig. 7.1. Heavy isotope build-up in natural uranium

some of the processes taking place in a reactor containing uranium-235 or uranium-238, or both, are given in Fig. 7.1; vertical arrows pointing upward represent beta decay, horizontal arrows pointing to the right are for (n, γ) reactions, and those pointing to the left are for $(n, 2n)$ reactions with fast neutrons. Those species which do not undergo beta decay are alpha active.

7.11. The conclusion of immediate interest to be drawn from Fig. 7.1 is that, in a reactor which operates for some time, various isotopes of uranium, neptunium, and plutonium, as well as of the heavier elements, americium (Am, atomic number 95), and curium (Cm, atomic number 96), may accumulate. This is referred to as a "heavy isotope build-up"; it may be particularly significant for power reactors which are expected to operate for a long time without refueling. In such reactors, it is possible that elements of still higher atomic number may be formed.

7.12. Some of the reactions which will occur in a reactor employing thorium-232 as the fertile material are shown in Fig. 7.2. The (n, γ) reactions are carried only as far as uranium-234, the point at which Fig. 7.1 commences. Since uranium-234 probably undergoes the $(n, 2n)$ process with fast neutrons to form

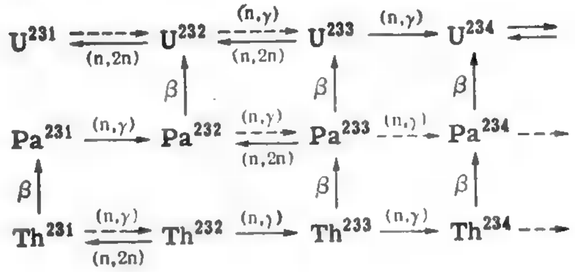


FIG. 7.2. Heavy isotope build-up in thorium

uranium-233, to some extent at least, the two charts together indicate possible heavy-element products in reactors based on uranium-233, uranium-235, or plutonium-239 as the fissionable species, with thorium-232 or uranium-238 as the fertile material.

7.13. It is evident that spent reactor fuel or a breeder blanket will contain a number of isotopes of high atomic number. The major element, i.e., uranium or thorium, will be present in large excess, but there will be appreciable amounts of products such as plutonium-239 or uranium-233, as well as traces of other elements. Some of these will represent intermediate stages of the regenerative process, e.g., neptunium and protactinium, whereas others will be unwanted (parasitic) capture products, including possibly americium and curium. In addition, the spent fuel will be associated with a large number of fission products in varying proportions, depending on their respective yields (§ 2.173). The problem of extracting the fissionable material and recovering the fuel in a reasonably pure state with a minimum of loss is thus a complex one requiring some knowledge of the chemistry of the heavy elements and of the fission products. Those aspects of the subject which are of importance to the separations processes will now be considered.

CHEMISTRY OF THE HEAVY ELEMENTS

LANTHANIDE AND ACTINIDE SERIES

7.14. It is well known that the sixth period of the periodic system of the elements contains a group of fifteen consecutive elements, generally known as the rare-earth series, having closely related chemical properties. This group owes its existence to special circumstances in connection with the arrangement of the orbital electrons (§ 1.46) of the various atoms, as indicated in Table 7.1.* The

TABLE 7.1. ELECTRONIC CONFIGURATION OF ELEMENTS IN SIXTH PERIOD

Element	At. No.	Numbers of Electrons								
		Shells 1, 2, 3	Shell 4				Shell 5			Shell 6
			4s	4p	4d	4f	5s	5p	5d	6s
Cesium	55	28	2	6	10	—	2	6	—	1
Barium	56	28	2	6	10	—	2	6	—	2
Lanthanum	57	28	2	6	10	—	2	6	1	2
Cerium	58	28	2	6	10	1	2	6	1	2
Praseodymium	59	28	2	6	10	2	2	6	1	2
.....	..	..	.	.	..	.	.	.	.	.
Lutetium	71	28	2	6	10	14	2	6	1	2

first two members of the sixth period, i.e., cesium and barium, have one and two electrons, respectively, in the outermost (sixth) shell, although the fourth and fifth shells are still incompletely filled. With the next element, lanthanum, the situation arises in which a more stable system results by the additional electron going into an inner (fifth) shell rather than into the outer (sixth) shell. In the succeeding fourteen elements, the greatest stability is achieved by the gradual filling up of a still-vacant (4*f*) sublevel in the fourth electron shell.

7.15. The fourteen 4*f* electrons are too deep within the atom to have much influence on the chemical properties of the elements; these properties are largely determined by the three outermost electrons, one in the fifth (5*d*) shell and the other two in the sixth (6*s*) shell. Consequently there exists a series of fifteen elements from lanthanum (atomic number 57) through lutetium (atomic number 71) having similar chemical properties. Although commonly known as the rare-earth elements, they are also referred to as the *lanthanide series*, after the name of the first member. Because they all have the three relatively loosely bound electrons, as stated above, the lanthanide elements form characteristic (hydrated) tripositive ions, e.g., La^{+3}, in solution. In addition, cerium forms a

* The letters *s*, *p*, *d*, *f* are the symbols used to designate subgroups within a main group (or shell) of electrons.

moderately stable tetrapositive ion, Ce^{+4}, since in this element a fourth electron can apparently be removed without too much difficulty.

7.16. It has long been expected that a group of related elements, similar to the lanthanides, should exist in the seventh period of the periodic system. Before the discovery of neptunium, plutonium, and the other transuranium elements, i.e., those with atomic numbers larger than that of uranium (92), the evidence for the occurrence of such a series was not decisive. But it is now apparent that there does exist, in the seventh period, a group of elements having definite resemblances to each other. By analogy with the rare-earth (or lanthanide) elements this group should commence with actinium, atomic number 89, and so it has been called the *actinide series*.

Properties of the Actinide Elements*

7.17. Actually, however, the marked similarities, which extend from lanthanum to lutetium in the lanthanide series, are not apparent for all the elements of the actinide series. For example, the stable tripositive (III) state, characteristic of the lanthanides, is not typical of all the actinide elements. Although actinium itself exhibits a stable (III) state, the next element, thorium, has a tetrapositive (IV) state in solution, and the succeeding element, protactinium, exists in a pentapositive (V) state. The most stable compounds of uranium in solution are those in the hexapositive (VI) state, but this element, like neptunium and plutonium, also exhibits (III), (IV), and (V) states.

7.18. The known positive valence, or oxidation, states of the elements of the actinide series are indicated in Table 7.2. An attempt is made to express the relative stabilities of the different states of each element. The most stable state

TABLE 7.2. RELATIVE STABILITIES OF OXIDATION STATES OF THE ACTINIDE ELEMENTS

Atomic No.	89	90	91	92	93	94	95	96	97	98	99	100
Element	Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	—	—
III	****	*	*?	**	**	***	****	****	****	****	****	****
IV		****	*	***	***	****	*		**			
V			****	*	****	**	*					
VI				****	***	***	*					

* G. T. Seaborg and J. J. Katz (Eds.), "The Actinide Elements," National Nuclear Energy Series, Div. IV, Vol. 14A, McGraw-Hill Book Co., Inc., 1954; M. C. Sneed, *et al.* (Eds.), "Comprehensive Inorganic Chemistry," Vol. 1, D. Van Nostrand Co., Inc., 1953, Chapter 3 (by G. T. Seaborg); M. W. Lister, *Quarterly Rev.* (Chem. Soc. London), **4**, 20 (1950).

is represented by four stars, and smaller numbers of stars are used for states of decreasing stability.

7.19. The situation with the actinide series is evidently more complicated than in the lanthanide series. In fact, there has been some controversy concerning the justification of the use of the term "actinide" to describe the series of elements in the seventh period. There is little doubt, however, that the gradual filling up of the incomplete inner (fifth and sixth) electron shells, in preference to the outermost (seventh) shell, does commence with actinium and continues through all the known heavier elements (Table 7.3).* The difference

TABLE 7.3. ELECTRONIC CONFIGURATIONS OF ELEMENTS IN SEVENTH PERIOD

Element	At. No.	Numbers of Electrons								
		Shells	Shell 5				Shell 6			Shell 7
		1, 2, 3, 4	5*s*	5*p*	5*d*	5*f*	6*s*	6*p*	6*d*	7*s*
Francium	87	60	2	6	10	—	2	6	—	1
Radium	88	60	2	6	10	—	2	6	—	2
Actinium	89	60	2	6	10	—	2	6	1	2
Thorium	90	60	2	6	10	—	2	6	2	2
Protactinium	91	60	2	6	10	1	2	6	2	2
Uranium	92	60	2	6	10	3	2	6	1	2
Neptunium	93	60	2	6	10	4	2	6	1	2
Plutonium	94	60	2	6	10	5	2	6	1	2

in behavior between the actinides, on the one hand, and the lanthanides, on the other hand, is ascribed to the expected relatively small difference between the energies of the electrons in the vacant fifth (5*f*) and sixth (6*d*) sublevels in the former series, as compared with the larger differences between the fourth (4*f*) and fifth (5*d*) vacant sublevels in the latter series. Consequently, whereas the loss of more than three electrons in positive-ion formation is rare in the lanthanide series, it is relatively common among the actinide elements.

7.20. In making a broad over-all examination of the compounds of the elements of highest atomic number, commencing with actinium (atomic number 89), it would appear that the concept of an actinide series is reasonable. The properties of uranium, neptunium, plutonium, americium, curium, berkelium, californium, and elements 99 and 100 † in the (III) state are similar, and they closely resemble those of actinium. The (IV) states of uranium, neptunium, plutonium, and americium resemble thorium, and, although this state of pro-

* There is some uncertainty concerning the electronic configurations of the actinide elements; the arrangements given here are intended primarily for illustrative purposes and may not be exact.

† At the time of publication of this book these elements had not been named.

tactinium is not commonly observed in solution, it does occur in some solid tetravalent compounds having structures similar to those formed by the other elements. There is, however, one important discrepancy which should be mentioned. The (V) states of uranium, neptunium, plutonium, and americium resemble each other, but in solution they apparently differ from the (V) state of protactinium. Nevertheless, the corresponding solid compounds do exhibit structural similarities.

7.21. If it were not for the existence of the several oxidation states of uranium, neptunium, and plutonium, and, in particular, the differences in their relative stabilities (Table 7.2), the separation of these elements from each other would probably have proved even more difficult than it is at present. Whereas the various actinide elements have similar chemical properties in a given valence state, these properties are frequently quite different in the different states. For example, the (III) and (IV) states can be precipitated from aqueous solution by means of fluoride, but not the (V) and (VI) states. Further, nitrates of the (IV) and (VI) states are appreciably soluble in certain organic solvents, but in the (III) state the nitrates are virtually insoluble in these liquids. Facts of this kind have been utilized in the development of processes for extracting and separating the closely related actinide elements.

7.22. Because of their importance in various aspects of the atomic energy program, the properties of the actinide elements and their compounds have been subjected to intensive study in recent years. As a result a great deal is now known of the chemistry of uranium and of some of the transuranium elements. For the present purpose, however, it will be sufficient to touch upon only those aspects necessary to permit an understanding of the basic principles employed for the extraction of fissionable species and for the recovery of fuel from the spent material resulting from the operation of reactors of various types. The first step will be to review briefly some of the essential properties of the various oxidation states.

The Tripositive (III) State

7.23. In the tripositive state in aqueous solution the actinide elements exist as ions having the general formula M^{+++} (or M^{+3}); these ions, like other ions in solution, are undoubtedly hydrated, being associated with several, probably six to eight or more, molecules of water.* In writing chemical equations, however, it is the general practice to omit the water of hydration, although its existence should be understood.

7.24. Except for the possibility of oxidation to higher positive valence states in certain cases, the tripositive actinide ions closely resemble those of the rare-

* All ions in aqueous solution are associated with a more or less definite number of molecules of water so that the ion is said to be "hydrated." Some of the water molecules may be bound chemically to the ion, e.g., $Cu(H_2O)_4^{+2}$, but there are almost invariably others, in addition, held by electrostatic forces.

earth elements, e.g., lanthanum. Like the latter, they form insoluble hydroxides, $M(OH)_3$, and fluorides, MF_3, which have little tendency to dissolve in the presence of excess of the corresponding anion, i.e., hydroxyl (OH^-) and fluoride (F^-) ions, respectively. The carbonates, phosphates, and oxalates are also insoluble. The tripositive chlorides, MCl_3, and nitrates, $M(NO_3)_3$, are soluble in water but do not dissolve to any appreciable extent in organic solvents. The tendency to undergo hydrolysis and to form complexes with various anions will be considered later (§ 7.30, *et seq.*).

The Tetrapositive (IV) State

7.25. In solution the tetrapositive ions of the actinide elements are in the form of M^{++++} (or M^{+4}) ions, together with a number of molecules of water of hydration. Apart from their oxidation-reduction behavior, they have similar chemical properties which are analogous to those of the rare-earth element cerium in the ceric (IV) state (see § 7.15). Since the tetrapositive state is characteristic of thorium in solution, the properties of this element may be regarded as typical of the (IV) state. The hydroxides, $M(OH)_4$, and the fluorides, MF_4, are insoluble and, like the corresponding (III) compounds, are not dissolved by excess of the anions. The tetrapositive phosphates and iodates are also insoluble. The chlorides and nitrates are soluble in water and are also appreciably soluble in certain oxygenated organic solvents, e.g., ethers, esters, and ketones. The tetrapositive ions undergo marked hydrolysis and complex formation, as will be seen below.

The Pentapositive (V) State

7.26. The pentapositive actinide ions differ from the (III) and (IV) states in being oxygenated rather than simple ions in aqueous solution. They are present almost exclusively, especially in acid solution, as MO_2^+ ions, presumably in a hydrated form.* As indicated above, the chemical properties of the pentapositive ions of uranium, neptunium, and plutonium in solution are quite different from those of protactinium (V). The latter probably tends to occur in solution as Pa^{+5} ions, but, since the corresponding base is weak, it is considerably hydrolyzed, except perhaps in strongly acidic media. The pentapositive (MO_2^+) salts of the actinide elements are soluble in water and probably also in some organic solvents, but they have, hitherto at least, played no part in separations processes.

7.27. Except for neptunium (and protactinium, of course), the (V) state is unstable with respect to the higher (VI) and lower (III) and (IV) oxidation states. Consequently, as will be shown below, spontaneous "disproportionation" occurs in solution, whereby two pentapositive ions essentially oxidize and

* The pentapositive condition of M in MO_2^+ can be understood by considering the latter to be made up of M^{+4} and two O^{-2} ions, i.e., $(O^{-2}M^{+4}O^{-2}) = MO_2^+$.

reduce one another to form a hexapositive and a tetrapositive ion. Where the tripositive ion is fairly stable, as it is for plutonium, this will also be formed.

The Hexapositive (VI) State

7.28. The hexapositive (uranyl) state of uranium, which is the most stable under ordinary conditions, has been well known for many years. Studies of the transuranium elements have served to show that, in their chemical properties in the (VI) state, other than those concerned with reduction to lower oxidation states, they are similar to uranium (VI). Hence the chemistry of the latter may be taken as typical of the hexapositive state. In acid solution the predominant ions have the formula MO_2^{++} (or MO_2^{+2}), although at low acidities hydrolysis results in the formation of $M_2O_5^{++}$, and $M_3O_8^{++}$, at least in uranium solutions.* The simple salts of MO_2^{+2} ions, e.g., chloride, fluoride, nitrate, and sulfate are nearly all appreciably soluble in water, and the nitrates, in particular, dissolve readily in oxygenated organic solvents. The considerable solubility of uranyl nitrate hexahydrate, $UO_2(NO_3)_2 \cdot 6H_2O$, in ordinary ether, for example, has been used in a commercial method for extracting and purifying this salt. One of the characteristic properties of the hexapositive state is the sparing solubility of the double salt of the acetates with sodium acetate; its general formula is $NaAc \cdot MO_2Ac_2$ or $Na(MO_2)Ac_3$, where Ac represents the acetate radical, $-C_2H_3O_2$.

7.29. The addition of alkali to a uranyl (UO_2^{+2}) solution results in the precipitation of uranate salts in which the uranium is present in the anion, i.e., in the negative ion. These precipitates are generally regarded as diuranates, e.g., $Na_2U_2O_7$ and $(NH_4)_2U_2O_7$, analogous to the familiar dichromates, but there is some doubt as to their exact composition. Neptunium and plutonium apparently behave in a similar manner, although the plutonium compounds (plutonates) are more soluble than the corresponding uranates.

Complex Formation

7.30. In many cases, a cation, especially if it has a high (positive) charge, is able to attach to itself one or more anions to produce what is called a "complex." Such complexes may have a net positive charge, e.g., ThF^{+3}, ThF_2^{+2}; they may be neutral, e.g., UO_2SO_4; or they may be negatively charged, e.g., $UO_2(SO_4)_2^{-2}$. The tendency to form complexes in any given case depends on both the cation and the anion concerned. For the elements of particular interest in separations chemistry it appears that, in first approximation at least, the formation of complexes involves electrostatic forces between the positive and negative ions. In these instances it is found, as a general rule, that the higher the charge on the

* The MO_2^{+2} ions may be thought of as arising from the combination of M^{+6} and two O^{-1} ions. The $M_2O_5^{+2}$ and $M_3O_8^{+2}$ may be considered as $MO_2^{+2} \cdot MO_3$ and $MO_2^{+2} \cdot 2MO_3$, respectively; MO_3 is the typical oxide of the hexapositive state.

cation and the smaller its size, the greater is the tendency to form complexes with a given anion.

7.31. In comparing the (IV) and (III) states of the actinide elements, it is known that the bare tetrapositive ions, i.e., M^{+4}, not only have a higher charge than the corresponding tripositive ions, i.e., M^{+3}, but they are also smaller in size.* Thus both charge and size indicate a greater tendency to form complexes by the tetrapositive ions, and such experimental observations as have been made lend support to this expectation. The complexes formed with a given anion by the tetrapositive actinide ions in solution are considerably more stable than those formed by the tripositive ions.

7.32. If the (V) and (VI) states existed as simple ions, i.e., M^{+5} and M^{+6}, they should exhibit an even greater tendency to form complexes. As stated above, the positive ions in solution for these oxidation states are MO_2^{+} and MO_2^{+2}, respectively, and this is, in fact, a manifestation of the strong complex-forming tendencies. However, the MO_2^{+} and MO_2^{+2} ions can themselves form relatively stable complexes with anions. In these cases the effective charge, which determines the relative stability of the complexes, is influenced both by the net charge, i.e., one and two, respectively, and also by the charge on the M atom, i.e., five and six, respectively. Thus, as far as complex formation is concerned, MO_2^{+} behaves as if it had a charge between 2 and 3, and MO_2^{+2} as if its charge were between 3 and 4. Consequently the tendency to form complexes by the four oxidation states of an actinide element decreases in the following order:

$$M^{+4} > MO_2^{+2} > M^{+3} > MO_2^{+}$$

for a given anion.

7.33. An examination of the effect of the anion on the complexes formed by a specific cation shows that there is a rough parallelism between the stability of the complex and the weakness of the acid formed by the anion. Thus, in considering the monobasic inorganic acids, hydrofluoric acid is one of the weakest in aqueous solution, and fluorides (F^-) tend to form the most stable complexes. On the other hand, the perchlorate ion (ClO_4^-), which is the anion of the strong perchloric acid, exhibits little tendency to form complexes. The anions of dibasic acids carry a double negative charge; consequently such ions as sulfate (SO_4^{-2}) and oxalate ($C_2O_4^{-2}$) usually form more stable complexes than do singly charged anions. The fluoride ion is an exception to the general rule, and the complex-forming tendencies of a number of common inorganic anions (in dilute acid solution) decrease approximately in the order,

$$F^- > PO_4^{-3} > SO_4^{-2} > NO_3^- > Cl^- > ClO_4^-,$$

for a given cation. The anions of weak organic acids often form exceptionally stable neutral complexes, some of which are of interest in connection with chemi-

* The radius of the Pu^{+4} ion in a crystal lattice is 0.86A, as compared with 1.01A for the Pu^{+3} ion.

cal separations. Further reference to such complexes and their stabilities will be made later.

7.34. It should be emphasized that the generalizations made above concerning the relative tendencies of various cations and anions to form complexes are to be regarded as a rough guide to the expected behavior. Although they are, on the whole, adequate for the purpose of the present chapter, it should be borne in mind that cases may arise in which complex formation is highly specific and is not related to the size and charge of the ions concerned.

7.35. In a solution containing a complex (or complexes) there will be an equilibrium between each complex, on the one hand, and the simple constituent ions, on the other hand. For example, in a solution containing tetrapositive M^{+4} ions and sulfate ions, the equilibrium

$$M^{+4} + SO_4^{-2} \rightleftharpoons MSO_4^{+2}$$

will be established. It is therefore apparent from the law of mass action that the proportion of the element M present as the complex ion MSO_4^{+2} will be increased by increasing the concentration of sulfate ions. At sufficiently high concentrations some of the M^{+4} ions may unite with two SO_4^{-2} ions forming the undissociated (or neutral) complex $M(SO_4)_2$. At still higher concentrations, it is possible that a negative complex may form; thus,

$$M^{+4} + 3SO_4^{-2} \rightleftharpoons M(SO_4)_3^{-2}.$$

In any given solution, several different complexes may be present, the relative amounts depending on the stabilities of the complexes and the over-all concentrations of the positive and negative ions. The greater the ratio of anion (negative ion) to cation (positive ion) concentration, the larger will be the proportion of the metal present in complex form and the greater the tendency to form species of increasing complexity.

Hydrolysis of Positive Ions

7.36. Hydrolysis, which refers to the interaction of ions with water, is, in certain respects, analogous to complex formation involving a weak acid. In the case of hydrolysis, water is the weak acid. Thus the first stage of hydrolysis of the M^{+4} ion would be represented by

$$M^{+4} + H_2O \rightleftharpoons M(OH)^{+3} + H^+,$$

as compared with the first stage of complex formation involving the weak acid HA, viz.,

$$M^{+4} + HA \rightleftharpoons MA^{+3} + H^+.$$

In accordance with the resemblance between hydrolysis and complex formation, it has been found in many cases that the tendency for a cation to undergo hydrolysis is greater the greater its charge and the smaller its radius. Thus, for the different oxidation states of an actinide element, the order of decreasing tendency to hydrolyze is

$$M^{+4} > MO_2^{+2} > M^{+3} > MO_2^{+}$$

at equivalent concentrations.

7.37. It may be noted, in passing, that the occurrence of the (V) and (VI) oxidation states as MO_2^{+} and MO_2^{+2} ions in aqueous solution is probably to be attributed to the strong tendency for the M^{+5} and M^{+6} ions to suffer hydrolysis. For example, the interaction of M^{+5} with water may be represented as

$$M^{+5} + 4H_2O \rightleftharpoons 4H^+ + M(OH)_4^{+}$$
$$\Downarrow\Uparrow$$
$$MO_2^{+} + 2H_2O$$

or

$$M^{+5} + 2H_2O \rightleftharpoons MO_2^{+} + 4H^+.$$

7.38. Since hydrolysis is always accompanied by the liberation of hydrogen ions, it is apparent that the extent to which a particular ion is hydrolyzed will depend on the acidity of the solution. At low acidities, i.e., at low hydrogen-ion concentrations, the equilibrium is shifted to the right and hydrolysis is increased. Conversely, in a sufficiently acid solution, i.e., at high hydrogen-ion concentrations, hydrolysis can be suppressed almost completely.

7.39. Because of the considerable hydrolysis undergone by highly charged positive ions in solutions of low acidity, such solutions tend to precipitate either a basic salt, i.e., a salt of a complex cation containing one or more hydroxyl groups such as MOH^{+3}, $M(OH)_2^{+2}$, or $M(OH)_3^{+}$, or the hydroxide of the metal, e.g., $M(OH)_4$, these representing increasing degrees of hydrolysis. Although the basic salts and hydroxide are usually insoluble in water, they frequently remain in colloidal solution, particularly at the lower concentrations. Such a colloid is readily formed in tetrapositive plutonium solutions of low acidity (0.1 molar or less). The colloid is sometimes referred to as "polymerized plutonium (IV)" or "plutonium (IV) polymer" since it consists of an aggregation of molecules containing tetrapositive plutonium. It is somewhat unreactive and requires heating with concentrated acid to restore it to true solution.

7.40. In the presence of anions which form complexes, the tendency for hydrolysis to occur at a given hydrogen-ion concentration is decreased. The reason is that, when complex formation is considerable, the concentration of free cations is greatly reduced, and the hydrolytic equilibrium, represented in § 7.36, is moved to the left. Alternatively, it may be considered that water molecules and complex-forming anions compete for the free cations in the solution; consequently formation of complexes suppresses the hydrolysis.

Oxidation-Reduction Properties of Actinide Ions

7.41. The fundamental stabilities, with respect to one another in solution, of the various oxidation states of an element are best represented by means of the

oxidation-reduction potentials of the different combinations. These are usually expressed, for simplicity, as the "formal" potentials. The formal potential is that acquired by an inert, e.g., platinum, electrode when inserted in a solution containing one formula weight of each of two oxidation states. For example, the formal oxidation-reduction potential of the (III)/(IV) system in a chloride solution would be the potential of an inert electrode in a solution containing one formula weight of MCl_3 and one of MCl_4 in a specified volume, usually 1 liter. When the oxidation-reduction potential depends on the hydrogen-ion concentration of the solution, as it does when the higher oxidation state is (V) or (VI) and the lower state is (III) or (IV), then the formal potential is that for a solution containing 1 mole of acid per liter.

7.42. If the salts (and the acid) were completely dissociated into simple ions with no complex formation and the activity coefficients were always the same, the formal potentials would be independent of the anions (or acid) present. However, these conditions are usually far from being satisfied in practice, and so formal potentials are given, when possible, for perchlorate solutions. Such systems most closely approach the ideal conditions for a high degree of dissociation into simple ions and relatively little tendency to form complexes (§ 7.33).

7.43. The formal potentials in volts of four combinations of the oxidation states of uranium, neptunium, plutonium, and americium are recorded in Table 7.4; the (III)/(IV) and (IV)/(VI) potentials are also plotted in Fig. 7.3. Most

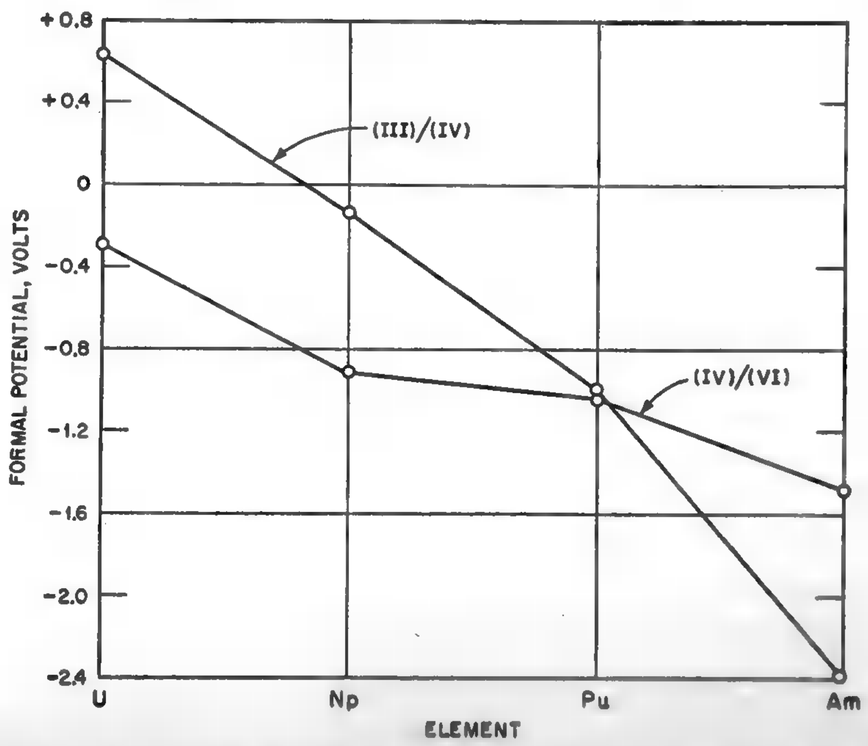


FIG. 7.3. Formal oxidation potentials for (III)/(IV) and (IV)/(VI) systems

of the values have been determined by direct measurement, but others have been calculated or estimated in various ways.* The more positive the potential, the greater the tendency for the lower state to become oxidized to the higher state, whereas the reverse is true the more negative the potential.

TABLE 7.4. FORMAL OXIDATION-REDUCTION POTENTIALS IN VOLTS

Element	(III)/(IV)	(IV)/(V)	(V)/(VI)	(IV)/(VI)
Uranium	+0.63	−0.61	−0.06	−0.33
Neptunium	−0.15	−0.74	−1.14	−0.94
Plutonium	−0.98	−1.15	−0.93	−1.04
Americium	−2.4	−1.4	−1.6	−1.5

7.44. An examination of the data in Table 7.4 and Fig. 7.3 reveals a number of important and interesting facts concerning the oxidation-reduction properties of the four elements. The steadily increasing negative values of the (III)/(IV) potentials indicate increasing stability of the (III) state with increasing atomic number of the element, from uranium to americium. The high positive value of the (III)/(IV) potential for uranium means that uranium (III) is so unstable with respect to uranium (IV) that it will be oxidized by water in aqueous solution. Reduction of uranium to the (III) state is therefore very difficult, and even when formed the solution of uranium (III) will reoxidize spontaneously. The small negative neptunium (III)/(IV) potential shows that, although neptunium (III) is somewhat more stable than uranium (III), it is nevertheless also easily oxidized in solution. In fact, exposure to air results in the ready oxidation of neptunium (III) to neptunium (IV).

7.45. The ease of oxidation from the (IV) to the (VI) state evidently decreases with increasing atomic number, as shown by the potentials in the last column of Table 7.4 (also Fig. 7.3). Thus, at one extreme, uranium (VI) is very stable and is difficult to reduce, whereas, at the other extreme, americium (VI) can be obtained only by very strong oxidation of the (III) or (IV) state. Uranium, neptunium, and plutonium can be oxidized to their (VI) states by means of a moderate oxidizing agent, such as hot bromate in acid solution. A fairly mild reducing agent, e.g., iodide ions, can then reduce the neptunium and plutonium to the (III) or (IV) state, but the uranium (VI) will be unaffected.

7.46. It is important to note, in connection with various separation processes, that, in a solution containing uranium (VI) and plutonium (III) or (IV), the uranium will not oxidize the plutonium or the plutonium reduce the uranium because of the small negative values of the uranium (IV)/(VI) and (V)/(VI) potentials. The data in Table 7.4 indicate, however, that uranium (VI) should be capable of oxidizing neptunium (III) to the (IV) state, that is to say, neptunium (III) should be able to reduce uranium (VI).

* It will be noted that the (IV)/(VI) potential is equal to ½[(IV)/(V) + (V)/(VI)].

7.47. Because of the similar values of the (IV)/(VI) potentials for neptunium and plutonium, it is apparent that neptunium (VI) and plutonium (VI) can be reduced to the (IV) state with approximately the same ease. But, owing to the closeness of the plutonium (IV)/(VI) and (III)/(IV) potentials, the reduction of this element will proceed to the (III) state if the conditions are suitable for the reduction of plutonium (VI). With neptunium, the reduction can easily be stopped at neptunium (IV) since a more powerful reducing agent is required to convert this to the (III) state.

Effect of Complex Formation

7.48. The reduction of plutonium (VI) can be stopped at the (IV) stage by taking advantage of the effect of complex formation; this will change the concentration of the simple ions present in solution and will thereby affect the oxidation-reduction potential. The latter is given approximately by the equation

$$E = E_f - \frac{2 \times 10^{-4} T}{n} \log \frac{[\text{higher oxidation state}]}{[\text{lower oxidation state}]}, \qquad (7.48.1)$$

where E_f is the formal potential (Table 7.4), T is the Kelvin temperature, n is the difference in the number of charges on the metal atom in the higher and lower oxidation states,* and the quantities in the brackets refer to the concentrations of the respective simple ions. Thus, if the higher oxidation state forms complexes so that the concentration of free ions is reduced, the oxidation-reduction potential, as given by equation (7.48.1), will become more positive. Alternatively, if the complex formation takes place preferentially with the lower oxidation state, the potential will become more negative.

7.49. As stated earlier, tetrapositive ions have a greater tendency to form complexes with anions than do tripositive ions under the same conditions. Hence the presence of complex-forming sulfate ions in a plutonium system will make the (III)/(IV) potential more positive so that the reduction of the (IV) to the (III) state will be more difficult. By adding sulfate to the solution and making a proper choice of reducing agent, it is thus possible to halt the reduction of plutonium (VI) at the (IV) stage with very little formation of plutonium (III).

Influence of Hydrogen-ion Concentration

7.50. When the oxidation or reduction process involves a change in the number of hydrogen ions, the potentials will be affected by the acidity of the solution. The (III)/(IV) and (V)/(VI) systems may be represented by

$$M^{+3} \rightleftharpoons M^{+4} + e^-$$

and

$$MO_2^{+} \rightleftharpoons MO_2^{+2} + e^-,$$

* The number of charges on the metal atom is the same as the number designating the particular oxidation state, i.e., 3, 4, 5, or 6 for the (III), (IV), (V), or (VI) state, respectively.

respectively, where e^- indicates a negative charge, i.e., an electron. The oxidation-reduction potentials are thus independent of pH, except in so far as the concentrations of the free ions may be affected by hydrolysis.

7.51. In the (IV)/(V) and (IV)/(VI) systems, however, hydrogen ions are concerned, as is evident from the equations,

$$M^{+4} + 2H_2O \rightleftharpoons MO_2^{+} + 4H^+ + e^-$$

and

$$M^{+4} + 2H_2O \rightleftharpoons MO_2^{+2} + 4H^+ + 2e^-,$$

so that the potentials will vary with the acidity. In each case the hydrogen ions are part of the higher oxidation state, and so the concentration of these ions will appear as a factor in the numerator of equation (7.48.1). It can be seen therefore that, for both (IV)/(V) and (V)/(VI) systems, decreasing the acidity, i.e., decreasing the hydrogen-ion concentration or increasing the pH, will make the oxidation-reduction potential more positive. Consequently reduction of either the (VI) or the (V) state to the (IV) state will be more difficult the less acid the solution. On the other hand, oxidation of the (IV) state will be facilitated by a decrease in the acidity. Because of the increasing tendency for hydrolysis and precipitation (or colloid formation) to occur, there is a practical limit to the extent to which the acidity may be decreased.

Rates of Oxidation-Reduction Reactions

7.52. Although the oxidation-reduction potentials indicate whether or not a particular process is possible under specified conditions, they provide no information concerning the rates at which the reactions will occur. A rough guide to this aspect of the problem is provided by considering whether a particular reaction requires the breaking and making of bonds or whether electron transfer only is involved. In the former case the reaction is likely to be slow, but in the latter case it will usually be rapid. As can be seen from the equations in § 7.50, in the oxidation of M^{+3} to M^{+4} or of MO_2^{+} to MO_2^{+2}, or in the corresponding reductions, no bonds are broken. Hence, if the reagent used for oxidation or reduction is one in which there is only a transfer of electrons, e.g., ceric, ferric, or argentic ions for oxidation, and ferrous or iodide ions for reduction, these reactions will probably occur rapidly.

7.53. Since oxidation of M^{+4} to MO_2^{+} or to MO_2^{+2} or the corresponding reductions require the breaking and making of bonds, in the water molecules as well as in the MO_2^{+} or MO_2^{+2} ions (§ 7.51), these processes will usually be slow. Advantage may be taken of the difference in oxidation or reduction rates in order to favor the temporary formation of one particular state of an element in preference to another.

Disproportionation Reactions

7.54. The possibility of *disproportionation*, i.e., simultaneous oxidation and reduction of the (IV) state into the (III) and (VI) states or of the (V) state into

the (IV) or (III) and (VI) states, can be readily determined from the oxidation-reduction potentials in Table 7.4. For the disproportionation of the (IV) state, written symbolically as,

$$3(\text{IV}) \rightleftharpoons 2(\text{III}) + (\text{VI})$$

or, more explicitly, as

$$3M^{+4} + 2H_2O \rightleftharpoons 2M^{+3} + MO_2^{+2} + 4H^+,$$

the so-called "disproportionation potential" is equal to the difference between the (IV)/(VI) and the (III)/(IV) potentials. Similarly, for the disproportionation of the (V) state, i.e.,

$$2(\text{V}) \rightleftharpoons (\text{IV}) + (\text{VI})$$

or

$$2MO_2^+ + 4H^+ \rightleftharpoons M^{+4} + MO_2^{+2} + 2H_2O,$$

the potential is the difference between the (V)/(VI) and (IV)/(V) oxidation-reduction potentials. The more positive the disproportionation potential, the greater should be the tendency for the process to occur.

7.55. The values of the disproportionation potentials of uranium, neptunium, and plutonium, calculated in the manner just indicated, are given in Table 7.5.

TABLE 7.5. DISPROPORTIONATION POTENTIALS IN VOLTS

Element	$3(\text{IV}) \rightleftharpoons 2(\text{III}) + (\text{VI})$	$2(\text{V}) \rightleftharpoons (\text{IV}) + (\text{VI})$
Uranium	−0.96	+0.55
Neptunium	−0.79	−0.40
Plutonium	−0.06	+0.22

It is apparent that uranium (IV) and neptunium (IV) will be relatively stable with respect to disproportionation, but plutonium (IV) should exhibit an appreciable tendency to undergo such reaction. It has been found experimentally that, upon standing at 25°C, a solution consisting initially of a pure plutonium (IV) salt in 0.5 molar hydrochloric acid contains 27.2 per cent of the (III) state, 58.4 per cent of the (IV) state, and 13.6 per cent of the (VI) state; the remaining 0.8 per cent or so is in the (V) state. With regard to the disproportionation of the (V) state, the negative potential for neptunium implies that pentapositive neptunium is much more stable than the corresponding forms of the other two elements. Under ordinary conditions, neptunium (V) appears to be the most stable state of this element in aqueous solution.

7.56. The effect of hydrogen-ion concentration on the disproportionation potential is determined by its effect on the oxidation-reduction potentials involved. However, it is obvious, by application of the law of mass action to the equilibria in § 7.54, that increase of hydrogen-ion concentration will suppress disproportionation of the (IV) state but will favor that of the (V) state. Thus

by decreasing the acidity, i.e., increasing the pH, the stability of the (V) state can be favored. Because of the increase in hydrolysis and the consequent possibility of causing precipitation or colloid formation, there is a limit to the increase in pH. There is thus an optimum stability range of pH for the pentapositive state; for uranium this has been estimated to be from pH 2 to 4.

7.57. In the foregoing treatment it has been supposed that the (V) state disproportionates into (IV) and (VI). There is, of course, the possibility that the (IV) state so formed will disproportionate to some extent into (III) and (VI). In the case of uranium (V), it is apparent from the data in Table 7.5 that disproportionation will occur largely into the (IV) and (VI) states. The disproportionation of plutonium (V), however, will yield (III), (IV), and (VI) states, leaving a small amount of the (V) state. The actual relative concentrations of the various states will depend on the character of the ionic medium, e.g., the nature and concentrations of the anions present.

7.58. Because disproportionation reactions all involve something more than electron transfer, it is expected that they will occur relatively slowly. Actually, since a large number of molecules or ionic species take part, e.g., $3M^{+4}$ and $2H_2O$ or $2MO_2^+$ and $4H^+$, these reactions must occur in stages, at least one of which may be slow.

7.59. It should be understood that the disproportionation reactions considered above and the reverse reactions represent aspects of the various equilibria existing among the ions of the four oxidation states that are present in solutions of uranium, neptunium, or plutonium. In principle all the possible ions, MO_2^{+2}, MO_2^+, M^{+4}, and M^{+3}, are always present, but the relative amounts at equilibrium are determined by the concentration of the solution, its acidity, and the presence of complex-forming anions and oxidizing or reducing agents. In this connection it may be pointed out that water can act in either of the latter capacities. In spite of the apparent complexity of the situation, it is nevertheless usually possible to adjust the conditions so that one particular oxidation state predominates over all others.

CHARACTERISTICS OF THE FISSION PRODUCTS

Relative Importance of Fission Products

7.60. It was seen in § 2.173 that the fission products are a mixture of more than 30 elements, from zinc (atomic number 30) to gadolinium (atomic number 64). Actually some 200 different isotopes have been detected, because most of the primary fission fragments are radioactive and undergo several stages of beta decay (§ 2.178). But from the chemical standpoint it is the nature of the elements present in the fission products that is important. The fact that most of them exist in several different isotopic forms is of no significance since the isotopes of a given element have essentially identical chemical properties. Any chemical process which removes one of the isotopes will remove all of them.

7.61. The considerable number of different elements in the fission products and the intense radioactivity of the mixture make the isolation and purification of the desired products, e.g., plutonium and uranium, a very difficult problem. This is accentuated by the great divergence in the amounts of the various species (see Fig. 2.25) and the abnormal behavior frequently exhibited by certain elements in very dilute solutions. Fortunately the situation is somewhat simplified in practice because, for reasons given below, fission products of very short or very long half life or which emit only beta radiations of fairly low energy can be largely ignored in the separation process. The same is true, for obvious reasons, for species which are formed in negligible yields; that is to say, they are the products of a very small proportion of fissions.

7.62. After removal from a reactor a spent fuel element is stored under water for a period, which may be as long as 100 days, before processing. This is generally referred to as the "cooling" period (see § 2.182). Its purpose is partly to permit the beta- and gamma-active neptunium-239, having a half life of 2.3 days, to decay almost completely to plutonium-239. Although most of the neptunium would probably be extracted with the plutonium in the separation process so that little of the latter element would be lost, the gamma activity of the product would be a serious hazard. It is consequently simpler to allow time for the neptunium to decay before treatment rather than after. During the cooling period, iodine-131, a fission product with a 8.0-day half life, which would be a radiation hazard if present in the gases from the processing plant, also undergoes considerable decay. At the same time, all short-lived fission products decay to such an extent that they are no longer significant as far as separations chemistry is concerned.

7.63. Within reasonable limits, fission products emitting beta particles of low energy, e.g., less than 1 Mev or so,* and no gamma radiation can be ignored since they do not represent an important hazard in the handling of the final product. Further, species with long half lives are not important since they decay (and emit radiation) at a very low rate. It is understood, of course, that isotopes having large capture cross sections for neutrons must not be present in appreciable amounts.

7.64. Based on the assumption that there is an ample cooling period before processing irradiated fuel elements, it is possible to draw certain conclusions concerning the importance of various fission products. It should be pointed out that the long cooling time means that there is a considerable hold-up of both fertile and fissionable material. It is possible that in the future it may become desirable or necessary, for economic reasons, to process reactor fuel after a short time interval. The circumstances will then be somewhat different since fission products of relatively short life will still be present. Nevertheless, the general

* Apart from the high-energy beta particles themselves, the bremsstrahlung accompanying their deceleration in passing through matter (§ 2.24) must not be overlooked.

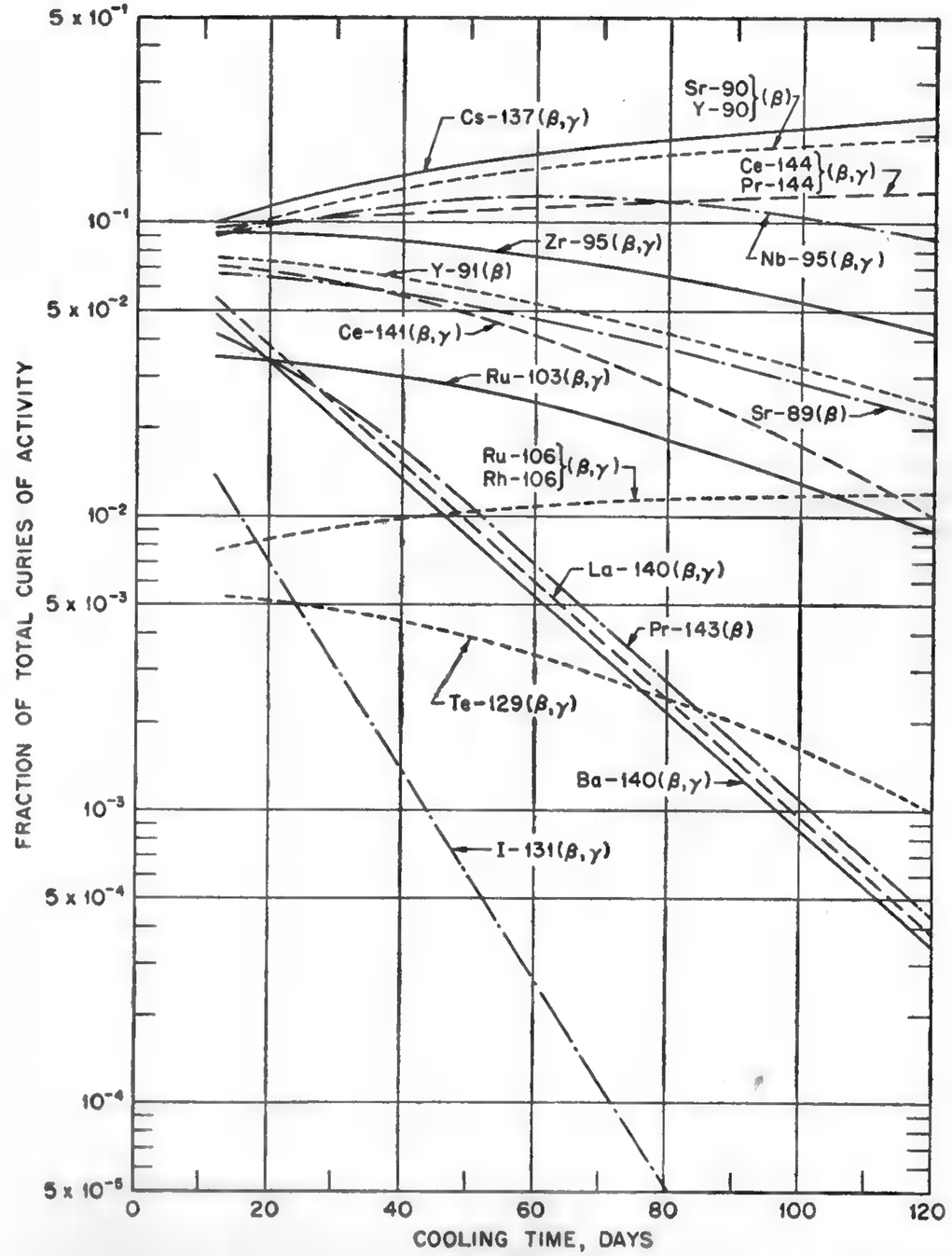


FIG. 7.4. Isotopes making major contributions to radioactivity of fission products

ideas expressed below will provide a basis for understanding the situation under different conditions.

7.65. Before considering the fission products from the standpoint of their chemistry, it is of interest to examine the curves in Fig. 7.4.* These show the major contributions to the total fission product radioactivity (in curies) made by a number of isotopes after various cooling times. The results are based on the supposition that the fuel elements have been in a reactor for an extended period so that at the time of shut-down an approximate condition of equilibrium has been attained. It will be evident from the figure that the isotopes of some twelve or thirteen elements are responsible for nearly all of the activity after 100 days or so of cooling.

Fission Products and the Periodic System

7.66. Since elements in the same group of the periodic system have similar chemical properties, it is convenient to consider the fission products according to the groups in which they appear. The treatment will be restricted to those isotopes formed in appreciable amounts.

Group 0

7.67. The gaseous fission products, krypton and xenon, belong to this group. These elements are chemically inactive and are expelled with the stack gases when the fuel element is dissolved. The elements of Group 0 are thus not important in the separations process chemistry.

Group I

7.68. This group is represented by rubidium and cesium in the A subgroup and by silver in the B subgroup, but, because of their short half lives or low yields, none of the isotopes of rubidium and silver need be considered here. Cesium, however, has two isotopes of long life. Cesium-135, formed with 5.9 per cent yield (§ 2.173), has a half life of 3×10^6 yr, but this is so long that the disintegration rate is negligible in comparison with that of cesium-137 (half life 33 yr) formed in 6.2 per cent yield. Nevertheless cesium is no problem in fuel processing because all the common salts are appreciably soluble in water but insoluble in organic liquids. Separation of cesium is therefore a simple matter.

Group II

7.69. Among the fission products there are strontium and barium in Group IIA and zinc and cadmium in Group IIB. Here again the members of the B subgroup are of no importance in processing because of the small yields or the short half lives of their isotopes. The alkaline-earth elements strontium and barium, however, have a number of isotopes of fairly long life formed in appreciable yields as a result of fission; they are strontium-89 (half life 53 days),

* See also, H. F. Hunter and N. E. Ballou, *Nucleonics*, **9**, No. 5, C-2 (1951).

strontium-90 (half life 28 yr), and barium-140 (half life 12.8 days). These three species, together with lanthanum-140, the decay product of barium-140, are responsible for as much as 21.7 per cent of the total fission product activity after 100 days of irradiation of a fuel element followed by 100 days of cooling.

7.70. The chemical properties of strontium and barium are similar except that, in general, salts of strontium are more soluble than the corresponding barium salts. Thus, in the presence of 1 to 2*M* nitric acid, barium sulfate is effectively precipitated completely, but strontium sulfate is only partially precipitated. Strontium phosphate does not precipitate in acid solution, although barium phosphate does to some extent. Similarly, whereas barium fluoride is completely carried down by a precipitate of lanthanum fluoride, strontium is carried only to a limited extent. In approximately neutral or alkaline solutions both strontium and barium can be precipitated as carbonate and oxalate.

Group III and the Lanthanides

7.71. Group IIIA is represented by gallium and indium, and Group IIIB by yttrium. Since the latter behaves chemically very much like the rare-earth elements, it is convenient to include the lanthanides with Group III, especially since lanthanum, at least, may be regarded as a member of the B subgroup. In this case it is the elements of Group IIIA which may be ignored from the standpoint of separation, and only those in the B subgroup that need to be considered.

7.72. Yttrium-91 (half life 61 days), having a fission yield of 5.4 per cent, is the main radioisotope of this element obtained in fission. There is also yttrium-90 (half life 61 hr) which results from the decay of strontium-90, but, since the latter has a half life of 28 yr (§ 7.69), only a small amount of yttrium-90 will be formed during a reasonable cooling period. In any event this isotope does not emit gamma radiation and so it is not a serious problem.

7.73. Eight rare-earth elements, namely, lanthanum, cerium, praseodymium, neodymium, promethium, samarium, europium, and gadolinium, are found among the fission products, but the last five need no further mention because of low fission yields or because of the very short or very long half lives of their isotopes.* Praseodymium may be of some significance for, although praseodymium-143 (half life 13.7 days), formed with a yield of 5.4 per cent, emits no gamma radiation, the energy of its beta radiation approaches 1 Mev, so that the bremsstrahlung may be important. There is a beta- and gamma-active isotope, praseodymium-144, with a half life of only 17.5 min, which is the decay product of cerium-144, referred to below. But, since the latter has a fairly long life, it is the chemistry of cerium, rather than of praseodymium, that is important in the separation process. Thus only lanthanum and cerium need be considered; of these, the latter is the more important, provided there has

* Certain isotopes of samarium, europium, and gadolinium, which make a negligible contribution to the radioactivity, have large cross sections for neutron absorption. They should consequently be removed from material to be used as reactor fuel.

been an appreciable cooling period, but the reverse is true for short periods, e.g., 40 days or less.

7.74. Cerium-141 (half life 33 days) and cerium-144 (half life 290 days), which decays to the active praseodymium-144, are found among the fission products, the yields being 6.2 and 4.9 per cent, respectively. Thus cerium is responsible for both short-term and long-term radioactivity. Assuming a 100-day irradiation time, cerium contributes 17 per cent of the total fission-product activity after 30 days of cooling, 15 per cent after 100 days, and 22 per cent after 2 yr. If the fuel is processed within 30 to 40 days after removal from a reactor, the most important rare-earth element, because of its highly penetrating gamma radiation, is lanthanum-140 (half life 40 hr), the decay product of barium-140.

7.75. Although yttrium does exhibit some differences in behavior from that of the rare-earth elements, they are, in a sense, differences of degree. For the present purpose it is the similarities which are more important, and the chemical properties mentioned below are those common to both yttrium and the lanthanides. The fluorides, phosphates, carbonates, and oxalates of these elements are insoluble in water, although yttrium carbonate and oxalate are fairly soluble in the presence of excess ammonium carbonate and oxalate, respectively. The fluorides do not dissolve in solutions of alkali fluorides or in dilute mineral acid containing hydrogen fluoride. The hydroxides, precipitated by alkali or ammonium hydroxide, are insoluble in excess of the reagent.

Group IV

7.76. Of the elements in Group IV, namely, germanium and tin in subgroup A and zirconium in subgroup B, only the latter is important in process chemistry. The main radioisotope in fission products is zirconium-95 (half life 65 days) formed in 6.4 per cent yield; both this isotope and its decay product, niobium-95 (half life 35 days), emit penetrating gamma radiation. For this reason, and also because of its very diverse chemical properties and its high yield, zirconium is one of the most troublesome elements in the treatment of reactor fuel.

7.77. In its characteristic (tetrapositive) form, zirconium exhibits amphoteric behavior, that is to say, the oxide (ZrO_2) has both basic and acidic properties. Three classes of salts are known: (1) normal Zr^{+4} compounds, e.g., $ZrCl_4$, which readily undergoes hydrolysis; (2) zirconyl compounds, e.g., $ZrOCl_2$; and (3) zirconates, e.g., Na_2ZrO_3 and $CaZrO_3$, in which the ZrO_2 behaves as an acid. Addition of alkali hydroxide to an acid solution of a zirconium salt results in the precipitation of zirconium hydroxide, $Zr(OH)_4$; this is insoluble in excess alkali and is even difficult to dissolve in acid unless precipitated in the cold. The alkaline carbonates precipitate Zr^{+4} as a basic carbonate, which is soluble in excess reagent but is precipitated on boiling. From acid solutions, phosphoric acid or phosphate precipitates Zr^{+4} completely as the zirconyl compound, $ZrO(H_2PO_4)_2$.

Group V

7.78. Two members of Group VA, namely, arsenic and antimony, appear among the fission products, but neither of these is important; in Group VB, however, there is niobium-95 (half life 35 days), the decay product of zirconium-95 (half life 65 days). Like its parent, niobium* is amphoteric in character. Its frequently uncertain chemical behavior, combined with its high yield and gamma activity, makes niobium one of the more difficult problems in separation processes.

Group VI

7.79. This group is represented by selenium and tellurium in subgroup A and by molybdenum in subgroup B. Because of their very long or very short half lives and low yields, none of the isotopes of selenium is significant. Of some, although not of major, importance is tellurium-129, produced in fission with a yield of 0.19 per cent. This isotope is formed in an excited (isomeric) state, half life 33.5 days, and the decay product, normal tellurium-129, has a short half life (72 min), emitting beta particles of high energy and also gamma radiation. The radioisotope molybdenum-99, with a fission yield of 6.2 per cent, has a half life of 67 hr. Consequently during a normal cooling period this will have decayed almost completely. However, because of its gamma activity, it might have to be considered if the cooling period were less than about 20 days.

Group VII

7.80. Among the fission products, bromine and iodine are in Group VIIA and technetium in Group VIIB. Iodine is the only one of these elements that is important in fuel treatment. Bromine has no long-lived radioisotopes, and the only isotope of technetium formed in appreciable yield, i.e., technetium-99, has such a long half life (2.12×10^5 yr) that its activity is negligible. The only important isotope of Group VII is iodine-131 (half life 8.0 days, yield 2.8 per cent). Its significance lies in the fact that iodine is likely to be evolved in the elemental state as a vapor during processing and would thus create an airborne radiation hazard. As indicated earlier (§ 7.62), much of the iodine decays during the cooling period, and most of the remainder is evolved with the gases accompanying the dissolution of the fuel elements.

Group VIII (Transition Group)

7.81. Although three of the transition-group elements, namely, ruthenium, rhodium, and palladium, result from fission, only ruthenium is important in separations chemistry. Rhodium-106 has a very short half life, namely, 30 sec, and palladium-107, with a half life of about 7×10^6 yr, is produced in very small yield. Thus both these isotopes can be ignored in the treatment of spent

* Although chemists now call this element "niobium," the alternative name "columbium" is still generally employed by metallurgists and engineers (see Chapter VIII).

fuel. Ruthenium-103 (half life 41 days) and ruthenium-106 (half life 1 yr) are present in appreciable amounts among the fission products. The beta activity of the latter isotope is quite weak, but its decay product, rhodium-106 (see above), emits highly energetic beta particles as well as some gamma radiation. Because of the very short half life of rhodium-106, its parent, ruthenium, is the determining element as far as separations chemistry is concerned.

7.82. On the whole, it is probably true to say that ruthenium is the most difficult element to separate from the desired products in fuel processing. The reason is that, in addition to having amphoteric properties, it exhibits several, possibly six, oxidation states. It will be seen later that the extraction of uranium and plutonium depends on certain changes in the oxidation states of these elements. Since ruthenium may well change its oxidation state at the same time, it is evident that its removal may present difficulties.

7.83. In acid solutions, ruthenium usually exists in the (III) or (IV) states, but in alkaline media it may occur in the (V), (VI), or (VII) states. When a ruthenium solution is strongly oxidized by a mixture of nitric and perchloric acid and is heated, the volatile oxide RuO_4, representing the (VIII) state, distills off. Ruthenium solutions are easily reduced to the elemental state, the metal plating out on solid surfaces. It is either this property or the tendency of a hydrolytic product to form a colloid (§ 7.39) that causes considerable unexpected trouble in the separation processes.

Summary

7.84. In reviewing the foregoing outline of the importance of the fission products, as far as processing spent fuel is concerned, it is seen that only nine or ten elements are of major significance. These elements are indicated below, together with the approximate weight percentage of each present in reactor fuel after 100 days of cooling following a long exposure in a reactor.

Group II.....	Strontium (3.8%)	Group IV.....	Zirconium (9.9%)
	Barium (4.0%)	Group V.....	Niobium (4.7%)
Group III.....	Yttrium (2.3%)	Group VI.....	Tellurium (1.2%)
	Lanthanum (4.0%)	Group VII.....	Iodine (0.6%)
	Cerium (10.1%)	Group VIII....	Ruthenium (5.0%)

7.85. It will be noted that these elements constitute, under the exposure and cooling conditions mentioned, something like 45 per cent by weight of the fission products. Among other elements which are present in significant amount, mention may be made of neodymium (10%), xenon (10%), cesium (10%), molybdenum (8%), praseodymium (4.5%), samarium (3%), and technetium (2.7%). For reasons given above, however, they either make a relatively small contribution to the radioactivity or are gaseous and so need not be considered in the present connection.

7.86. Of the elements tabulated, iodine presents no serious difficulties in separations chemistry. Part of this element is volatilized when the fuel is dissolved and the remainder, like cesium, stays in solution in most precipitation reactions so that it can be readily separated.* The complex problem of the removal of over 30 elements from spent fuel, in order to recover either fertile or fissionable material, is thus greatly simplified. There are only nine or ten elements that are at all significant, and these fall into a few groups, in each of which the chemical properties are similar. Traces of praseodymium, resulting from the decay of cerium, will behave like the elements in Group III. From what has been indicated earlier, it will be apparent that it is only the elements zirconium, niobium, and ruthenium that present real difficulties in the treatment of spent reactor fuel.

Composition and Radioactivity of Spent Fuel

7.87. It was seen in § 1.87 that the fission of 1 gram of uranium-235 (or other fissionable species) is accompanied by the liberation of 1 megawatt-day of energy. Hence the production of this amount of energy in a reactor will result in the formation of roughly 1 gram of fission products. Suppose the Brookhaven (natural uranium) reactor operates at a power level of 28 megawatts; then after one year of full operation there will have been formed (28) (365) = 10,220 grams, i.e., about 23 lb, of fission products. This will be present in about 50 tons of uranium, so that the fission products will constitute about 0.023 per cent by weight of the spent fuel element.

7.88. Assuming a conversion efficiency of uranium-238 to plutonium-239 of 100 per cent, so that every nucleus of uranium-235 undergoing fission is replaced by one of plutonium-239, the amount of the latter formed after one full year of operation of the Brookhaven reactor is also approximately 23 lb. The small quantity of plutonium destroyed by fission and nonfission capture reactions may be neglected. The composition of the spent fuel will thus be roughly as follows:

Uranium (all isotopes).....	99.954%
Plutonium-239.....	0.023
Fission products.....	0.023

These figures are given to provide some idea of the magnitude of the problem involved in the recovery and purification of fertile and fissionable materials in the processing of spent reactor fuel. Each ton of such fuel, according to the foregoing considerations, will contain about 0.46 lb of plutonium, 0.46 lb of fission products, and just over 1999 lb of uranium. The proportion of uranium-235 in the latter will have been reduced from the normal value of 0.71 to slightly

* Nevertheless, the presence of iodine among the fission products cannot be ignored; it may react with the organic solvent used in some separation processes (§ 7.154) and, in any event, the iodine volatilized when the fuel is dissolved can cause difficulties.

over 0.68 per cent, partly by fission and partly by conversion to uranium-236 as a result of neutron capture.

7.89. The total radioactivity of the spent fuel can be obtained from Fig. 7.5, which gives the activity of the fission products in terms of curies, as a function of cooling time, for various periods, T_0, of operation of a natural-uranium reactor, at a power level of one watt.* In any particular case the actual activity can be determined by multiplying the value obtained from Fig. 7.5 by the actual reactor power in watts.

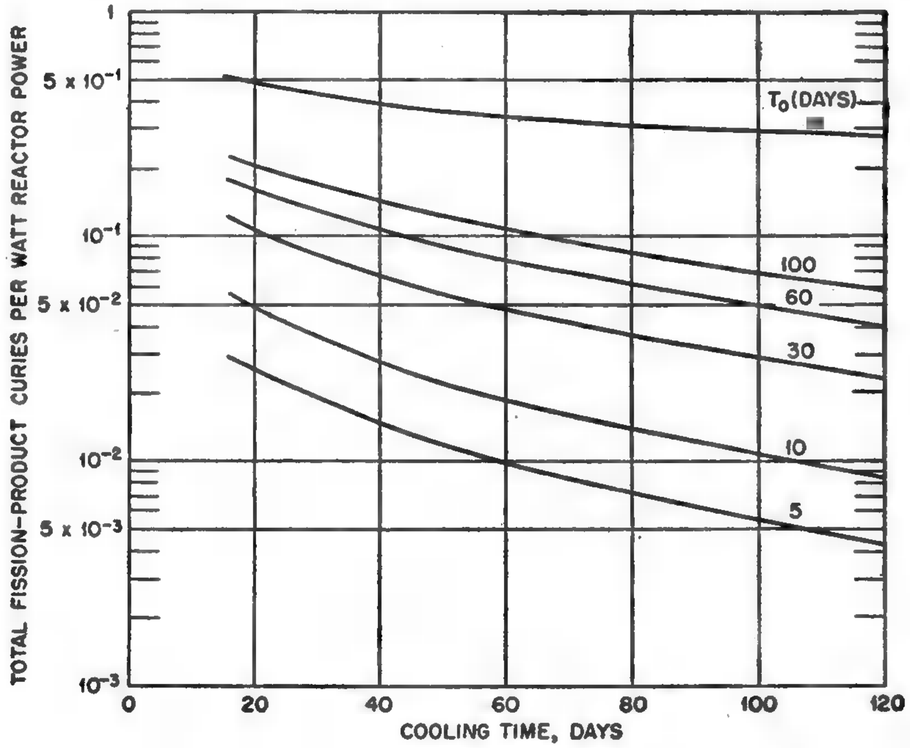


FIG. 7.5. Activity of fission products in natural-uranium fuel following T_0 days exposure in reactor

7.90. For an exposure of 365 days, the radioactivity after 100 days of cooling is estimated from the figure to be about 0.17 curie per watt. In the case of the Brookhaven reactor, for which the power level has been taken as 28 megawatts, i.e., 28×10^6 watts, the activity would thus be 4.8×10^6 curies. If the total amount of fuel present is 50 tons, this means an activity of 48 curies/lb of spent fuel. The proportion of this total contributed by any individual fission product can be obtained, if required, by means of Fig. 7.4.

7.91. It should not be concluded, on the basis of these calculations, that all the fuel elements in a reactor are discharged for processing at one time. It is

* See also, R. L. Koontz and A. A. Jarrett, *Nucleonics*, 12, No. 6, 26 (1954).

the general practice, where possible, to adopt a definite schedule for discharging and replacing a certain proportion only of the fuel elements. This procedure has the advantage of maintaining the over-all reactivity, since fresh fuel is introduced into the reactor at regular intervals. If this were not done, the reactivity would decline steadily over the whole period of operation. The calculations of fuel composition and reactivity made above are, nevertheless, applicable provided the exposure is the same as was assumed there, namely, $(365)\ (28 \times 10^6)$ watt-days/50 tons of fuel, i.e., 1.02×10^5 watt-days/lb.

SEPARATIONS PROCESSES: PRECIPITATION METHODS

General Principles*

7.92. Four main separations processes have been proposed in connection with the treatment of irradiated reactor fuel. These are (1) precipitation, (2) adsorption (or sorption), (3) solvent extraction, and (4) volatilization. The basic principles underlying these methods for separating elements from each other will be considered, and some indication will be given of their possible use for the separation of plutonium and uranium from the fission products.

7.93. Precipitation methods have long been used by chemists, both in the laboratory and on the large scale, for the separation of elements or for the removal of impurities from a desired product. In the extraction of plutonium from spent fuel a difficulty arises because of the small proportions of plutonium and of fission products present, as compared with that of uranium. The volumes of solution, at least in the early stages of the separations process, are so large that removal of the trace amounts of plutonium or of fission products by direct precipitation would not be practical. The problem can be solved by the use of a suitable "carrier." This is a substance added in appreciable amount to a solution of a trace element, which the latter will follow in a given chemical procedure. In the case under consideration, the plutonium (or the fission products) could be precipitated with a suitable carrier, and the resulting macroscopic precipitate could then be removed in the usual way, by filtration or centrifugation. If the precipitate contained the desired product, e.g., plutonium, a method would have to be found for removing it from the carrier. How this is done will be described later.

Carrying by Solid-solution Formation

7.94. Several different mechanisms have been proposed to explain the carrying of trace elements by precipitates. These mechanisms are not mutually exclusive and one, two, or more may be operative in any particular instance. The simplest, ideal type of carrying arises when the trace element is incorporated into the crystal lattice of the carrier precipitate, a solid solution (or mixed

* See A. C. Wahl and N. A. Bonner (Eds.), "Radioactivity Applied to Chemistry," John Wiley & Sons, Inc., 1951, Chapter 6 (by N. A. Bonner and M. Kahn).

crystal) being formed. This will occur, in general, when the precipitated salt of the trace element and of the carrier are isomorphous, i.e., they have the same crystalline form, and the ions of the two elements are not greatly different in size. Probable examples of this type of mechanism are the carrying by lanthanum fluoride (LaF_3) of rare-earth fluorides (MF_3), and possibly also of plutonium (III) fluoride.

7.95. A number of anomalous cases of carrying by formation of solid solutions are known; thus strontium fluoride (SrF_2) and lanthanum fluoride (LaF_3) coprecipitate in this manner, presumably because the Sr^{+2} and La^{+3} ions are similar in size. The resulting lattice is of the "defect" type since some of the points usually occupied by the fluoride ion in LaF_3 will be vacant when SrF_2 is present. It has been suggested that the carrying of plutonium tetrafluoride (PuF_4) by lanthanum fluoride is also due to anomalous isomorphism, although the Pu^{+4} ion (crystal radius 0.86 A) is considerably smaller than the La^{+3} ion (crystal radius 1.04 A). However, there is evidence that a sparingly soluble double salt $2LaF_3 \cdot PuF_4$, is formed, and this may be partly responsible for the carrying.

Nernst-Berthelot Distribution Law

7.96. If the solid-solution precipitate were formed instantaneously or if sufficient time were allowed for the precipitate to attain complete equilibrium with the (liquid) solution, it is probable that the trace element would be uniformly distributed throughout the solid. In this event the trace element is divided in a definite manner between the precipitate and the supernatant solution according to the Nernst-Berthelot distribution law,* i.e.,

$$\frac{\text{Conc. of trace element in precipitate}}{\text{Conc. remaining in solution}} = \text{constant } (k_t),$$

irrespective of the initial amount of trace element in the solution. A similar relationship should hold for the carrier, so that

$$\frac{\text{Conc. of carrier element in precipitate}}{\text{Conc. remaining in solution}} = \text{constant } (k_c).$$

Upon dividing the first of these two equations by the second, the result is

$$\frac{\text{Ratio of trace element to carrier in precipitate}}{\text{Ratio of trace element to carrier in solution}} = D, \qquad (7.96.1)$$

where the constant D, called the *homogeneous distribution coefficient*, is a property of the particular substances, not merely the elements, which are precipitated. Thus when plutonium trifluoride is carried by lanthanum fluoride, the distribution coefficient at a given temperature is a property of these particular fluorides, as well as of the solution from which precipitation takes place.

* The assumption is made here that both solid and liquid solutions of the trace elements behave ideally.

7.97. For good carrying it is desirable that most of the trace element should go into the precipitate with only a small proportion remaining in solution. In other words, it is necessary for the distribution coefficient D to be as large as possible; that is to say, the constant k_t should be large with respect to k_c. This condition can be realized, in general, if the compound of the carrier which is precipitated is more soluble in the aqueous solution than is the corresponding compound of the trace element. For example, lead sulfate is somewhat more soluble in water than barium sulfate, and consequently lead sulfate is a good carrier for barium.* On the other hand, barium sulfate would not be expected to be as good a carrier for the removal of lead from solution as is lead sulfate for barium.

7.98. In actual practice, the usual situation is that the precipitate is neither formed instantaneously nor is it permitted to stand for a sufficient length of time for complete equilibrium to be attained with the solution. An intermediate type of situation, which can be treated theoretically, is to suppose that the precipitate consists of crystals which steadily increase in size during the course of precipitation, equilibrium being continuously maintained between the growing outer layer of the crystal and the existing solution. Since the composition of the solution changes continuously during the course of precipitation, so also will that of the growing crystal surface. Thus each crystal of precipitate will represent a solid solution of continuously varying composition from the interior to the outer surface.

7.99. In this case, provided the composition of the precipitate does not change after its formation, it can be shown that the fraction, f_t, of the initial amount of the trace element which has been precipitated is related to the corresponding fraction, f_c, of the carrier element by

$$\log (1 - f_t) = D \log (1 - f_c), \qquad (7.99.1)$$

where D is the homogeneous distribution coefficient defined by equation (7.96.1). This expression is a form of what is called the Doerner-Hoskins (or logarithmic) distribution law.

7.100. It is evident from equation (7.99.1) that for efficient carrying, i.e., for f_t, the fraction of trace element precipitated, to be large for a given value of f_c, the homogeneous distribution coefficient, D, should also be large. Qualitatively this is the same result as was derived for the case of the formation of a uniform solid solution. The general conclusions drawn above for the simple case are thus also applicable in the present instance.

7.101. Mention should be made of the effect of complex-forming anions in the solution, which, as a general rule, tend to increase the solubility of the precipitate. It will be apparent that when the carrier element forms complexes in solution, but the trace element does not, the carrying will be enhanced. In these

* This is true even though the precipitate is not necessarily a homogeneous, i.e., uniform, solid solution, as postulated above (cf. ¶ 7.100).

circumstances the solubility of the carrier precipitate is increased, so that k_c will tend to decrease relative to k_t, thereby increasing the homogeneous distribution coefficient. On the other hand, if the trace element forms such complexes preferentially the extent of carrying will be decreased.

Carrying by Adsorption

7.102. Many precipitates, especially when freshly formed, have large surface areas and so appreciable amounts of trace elements may be carried by adsorption.* In these circumstances there is no obvious relation between the carrier precipitate and the material carried. Thus barium (II), zirconium (IV), and protactinium (V) are carried by lanthanum (III) fluoride from acid solutions, presumably by adsorption in each case. It appears, as a rough general rule, that highly charged positive ions are more readily adsorbed than those of lower charge. This may be due, in part at least, to hydrolysis and the consequent formation of colloidal solutions by the ions of high charge (§ 7.39).

7.103. Frequently the carrying can be improved by having excess of the anion, i.e., the ion of the acid, in the solution. For example, thorium-B, an isotope of lead, is carried much more efficiently by barium sulfate when the solution contains excess of sulfate ions. These ions are probably adsorbed by the barium sulfate, and the resulting negatively charged precipitate then attracts, and so carries, the positive thorium-B ions.

7.104. Because of this effect of the anions, the amount of a trace element adsorbed by a given precipitate frequently depends on the order in which the reagents are added. Thus, in the case just considered, better carrying would be expected if the solution of carrier and trace element were added to the sulfate (or sulfuric acid) solution than if the opposite order were employed. In the former instance the precipitate is always formed in the presence of excess of sulfate ion, so that adsorption of the trace element is favored.†

7.105. Apart from the effect of valence, mentioned above, and the fact that positive ions forming the least soluble compounds with the precipitating ion are usually, although not always, carried best (Fajans's rule), adsorption is a general phenomenon. As a result of its nonspecific character, there are some interesting consequences. In the first place a precipitate with a very large surface area, e.g., manganese dioxide or ferric hydroxide, is sometimes able to carry down with it a number of different trace elements from solution. Such a precipitate is then referred to as a *scavenger*.

* A distinction is made between *ad*sorption, in which a substance is held or carried on the surface only, and *ab*sorption, in which the substance is distributed throughout the whole of the absorber. In many cases it is not possible to distinguish between adsorption and absorption, and the general term *sorption*, without a prefix, is then used.

† The mixing of solutions in this order is sometimes called a *reverse strike*. When the precipitating ion, e.g., sulfate in the case under consideration, is added to the solution of carrier and trace element, it is called a *direct strike*. These terms are used irrespective of the method of carrying.

7.106. Another result of the nonspecific nature of carrying by adsorption is that carrying of trace elements can often be prevented by adding to the solution a fairly large amount of an indifferent substance which is well adsorbed by the precipitate. By occupying most of the surface, this added substance, called a *holdback carrier*, greatly decreases the amount of the trace element which is carried. The presence of zirconium, for example, appreciably decreases the proportion of protactinium carried by lanthanum as fluoride from a hydrofluoric acid solution.

7.107. Carrying by adsorption is obviously greatly dependent on the conditions existing in solution. Reference has been made above to the effect of the order of adding the reagents and to the influence of certain indifferent substances. Further, the concentrations of the trace, carrier, and precipitating ions, the rate of mixing of the solutions, and the temperature will all affect the quantity of trace element adsorbed by the carrier. The nature of the anions present will also have some influence, especially if they form complexes with the trace elements; this will have the effect of decreasing the amount carried.

7.108. For these and other reasons, it is not feasible to develop a quantitative expression for the amount of the trace element carried. However, from a knowledge of adsorption phenomena, it is possible to draw the following general conclusions. First, the *amount* of the trace element carried will increase with its concentration in the solution, but the *fraction* of the initial amount that is carried by the precipitate will be greater in the more dilute solutions. Since the trace element is inevitably present at low concentration, the circumstances are favorable to good carrying. Second, the quantity of trace element carried from a given solution increases in proportion to the mass of the precipitate formed. It is therefore desirable that a conveniently large amount of carrier be precipitated.

7.109. Although, as already stated, the amount of trace element adsorbed generally increases as its concentration in the solution is increased, a point may be reached when the surface of the carrier precipitate is saturated with the trace element. The amount of the latter carried will then be the maximum possible for the given quantity of precipitate under the existing conditions. In order to carry more of the trace element it would then be necessary to increase the proportion of carrier to trace element in the solution before precipitation. The effectiveness of a given carrier for a particular trace element will thus be determined by the relative concentrations. Difficulties arising in a particular situation may be overcome by changing the concentration or, if necessary, the nature of the carrier. Such problems are to be expected when carrying is by adsorption although they frequently arise when other mechanisms are operative.

Other Types of Carrying

7.110. Formation of solid solutions and adsorption represent the most important carrying mechanisms, but there are other types of carrying which do not

fall into these categories. One of them, called *internal adsorption*, is manifested by nonuniform distribution of the trace element within the carrier precipitate. This is attributed to adsorption of the trace element by the changing surface of the precipitate as the minute crystals increase in size. Since carrying by internal adsorption exclusively is not common, it need not be discussed further here. It should be noted in this connection that most cases of carrying probably involve two or more different mechanisms, although one type is usually predominant.

Applications of Precipitation Processes

7.111. Several precipitation methods for the separation of plutonium from spent reactor fuel have been worked out. Most of them depend on the fact that the simple salts of the hexapositive state of uranium and plutonium are usually soluble in water (§ 7.28), whereas there are a number of compounds of the tetrapositive state, e.g., fluoride, phosphate, oxalate, and iodate, which are insoluble (§ 7.25). Consequently plutonium can be separated from uranium by precipitation as one of these salts from a solution containing uranium (VI) and plutonium (IV) nitrates. In view of the relatively low concentration of plutonium in the solution, the use of a carrier is essential. Among those which have been considered is lanthanum because, as stated earlier, lanthanum fluoride is a good carrier for plutonium.

7.112. Some of the fission products, notably the rare-earth elements, will be carried down with the plutonium, and so they must be separated from the latter. This can be achieved by dissolving the carrier precipitate of lanthanum fluoride,

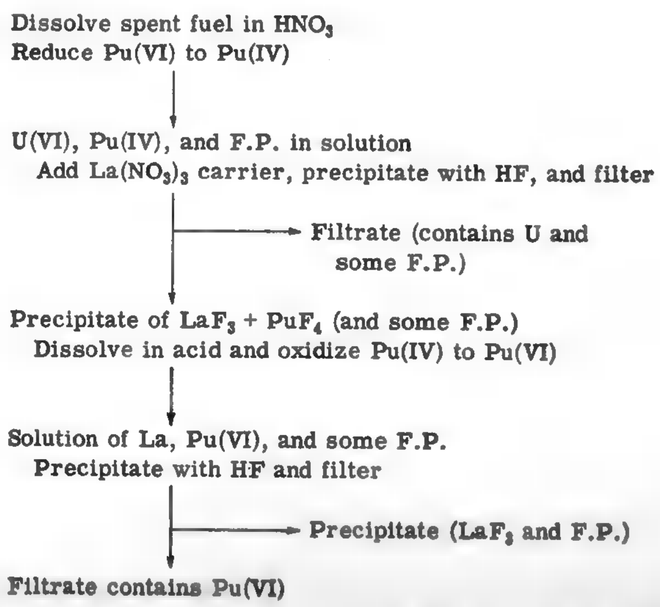


Fig. 7.6. Separation of plutonium by precipitation with carrier

containing the plutonium, and some impurities, in a suitable acid, oxidizing the plutonium to the (VI) state, and then reprecipitating the carrier. Since the simple salts of hexapositive plutonium are soluble, the precipitate will now carry only the fission product impurities, leaving the plutonium in solution (Fig. 7.6).*

7.113. In general, one stage of purification is not sufficient, and so the procedure is repeated. The plutonium is reduced to the (IV) state and once more precipitated with a carrier. The precipitate is dissolved, the plutonium oxidized to the (VI) state, and the carrier again reprecipitated. The residual solution will contain plutonium (VI) in relatively pure form. Zirconium (IV) and niobium (V) are difficult to remove since they are usually incompletely carried by the precipitate. This is probably because the highly charged ions of these elements readily undergo hydrolysis, and the products tend to remain in colloidal solution.

SEPARATIONS BY SORPTION

Molecular Sorption

7.114. Two different types of sorption phenomena have been applied in the separation of cations from one another. The first, and less important, involves a mechanism similar to that of adsorption carrying by precipitates. It may be regarded as *molecular* sorption, i.e., both positive and negative ions of a salt are sorbed, to distinguish it from the *ionic* sorption, to be considered below, in which either positive or negative ions, but not both, are taken up from solution.

7.115. Molecular sorption is based on the fact that certain solids, notably oxides, such as silica gel, alumina, and titania, can be prepared in a form in which they have very large surface areas per unit mass. As a result these substances, called sorbents, are able to take up considerable amounts of various electrolytes, as well as nonelectrolytes, from solution. Although the sorption is not very selective, except possibly for the valence effect mentioned in § 7.102, nevertheless, some substances will be taken up by the sorbent more readily than others present in the same solution. This may make possible a partial (or complete) separation of certain ions.

Ion-exchange Separations†

7.116. From the standpoint of nuclear reactor chemistry, at least, a more important phenomenon, which is a kind of sorption, is *ion exchange*. Certain solids, called *ion exchangers*, although themselves insoluble in water, are able to

* J. F. Flagg and E. L. Zebroski, Report AECU-1962; *Scientific American*, **187**, No. 1, 62 (1952).

† F. C. Nachod (Ed.), "Ion Exchange: Theory and Application," Academic Press, Inc., 1949; R. Kunin and R. J. Myers, "Ion Exchange Resins," John Wiley & Sons, Inc., 1950; J. F. Duncan and B. A. J. Lister, *Quarterly Rev.* (Chem. Soc. London), **2**, 307 (1948).

exchange either positive or negative ions with ions of the same charge present in an aqueous solution. Ion-exchange materials of various types are known, but those in most general use are based on synthetic resins, consisting of insoluble organic polymers of high molecular weight. These are called ion-exchange resins.

7.117. An ion-exchange resin consists, in general, of a loosely crossed-linked, polymerized organic structure to which is attached a number of active (or functional) groups. In cation-exchange resins the functional group is acidic in character, capable of splitting off a hydrogen ion; the "weak acid" resins usually contain carboxylic (—COOH) or phenolic (—OH) groups and the "strong acid" resins the sulfonic acid (—SO_3H) group. In anion-exchange resins, on the other hand, the active group is basic. In the "weak base" resins this is an amino (—NX_2) or an imino (=NX) group, where X may be hydrogen or an organic radical, but in those of the "strong base" type it is a quaternary ammonium (—NX_3^+) group, where X is an organic radical.

7.118. Most of the work on ion-exchange resins hitherto has been with those of the cationic type because they are easier to make and are more stable to various chemical reagents. Among the resins which have been widely used are the Dowex and Amberlite products; they are available in a variety of cationic and anionic types. In Dowex-50 and Amberlite IR-120, for example, the functional group is the strongly acidic sulfonic acid group. These resins are effective over a wide range of hydrogen-ion concentrations and have high capacities for cation exchange. They are relatively stable to acids, alkalis, and oxidizing and reducing agents even at 100°C.

7.119. The ion-exchange separation technique is particularly applicable to dilute solutions. It can be used for such purposes as (1) removal of desired or undesired substances present in solution, (2) concentration (or volume reduction) of solutions, (3) metathesis, i.e., replacement of one ion by another, and (4) separation of ions from one another. From the engineering standpoint ion exchange is a diffusion operation involving the transfer of ions from one phase to another in a liquid-solid system. The process is generally carried out with a fixed resin bed through which the solution percolates. The conditions are such that unsteady-state diffusion theory is applicable, the mathematical treatment being similar to that used in connection with batch distillation.

General Principles of Ion Exchange

7.120. When a solution is brought in contact with an ion-exchange resin, an interchange will generally occur between the ions in solution and the mobile ions of the same sign from the resin. For example, a cationic resin, represented by H—R, where R is the insoluble resin and H is the ionizable hydrogen which is part of the functional (acidic) group, will exchange with positive ions, M^{+n}. Thus, the equilibrium

$$n\ \text{H—R (resin)} + \text{M}^{+n}\ \text{(soln.)} \rightleftharpoons \text{M—R}_n\ \text{(resin)} + n\ \text{H}^+\ \text{(soln.)}$$

is established, so that part of the resin is converted into $M—R_n$ while hydrogen ions are set free in the solution. The amount of hydrogen ions liberated is equivalent to the quantity of M^{+n} ions retained by the resin, so that electrical neutrality is maintained. Similarly, for an anionic resin in the chloride form, i.e., R—Cl, the exchange with anions, A^{-n}, can be represented by

$$n\ \text{R—Cl (resin)} + \text{A}^{-n}\ \text{(soln.)} \rightleftharpoons \text{R}_n\text{—A (resin)} + n\ \text{Cl}^-\ \text{(soln.)},$$

so that a portion of the A^{-n} ions are retained by the resin while an equivalent amount of chloride ions is liberated in solution.

7.121. The amount of any particular ion remaining on a given quantity of an ion-exchanger resin increases with the concentration of the ions in solution. However, there is a limit to what is called the "capacity" of the resin; this varies with the nature of the resin, but is generally on the order of 2 to 5 gram equivalents per kilogram of resin.

7.122. The ability of a resin to retain (or its affinity for) different ions, under specified conditions, is determined by two factors; these are (1) the charge on the ion and (2) the size of the hydrated ion as it exists in solution. The greater the charge carried by the ion, either positive or negative, according to circumstances, and the smaller the radius of the ion in the hydrated form the more strongly will the ion be retained by a given resin. For the ions Th^{+4}, La^{+3}, Ba^{+2}, and Na^+, for example, the order of decreasing retention by a given resin from equivalent solutions, i.e., the order of decreasing affinity, is usually

$$\text{Th}^{+4} > \text{La}^{+3} > \text{Ba}^{+2} > \text{Na}^+.$$

The order of retention for the alkali metal series of univalent ions is

$$\text{Cs}^+ > \text{Rb}^+ > \text{K}^+ > \text{Na}^+ > \text{Li}^+,$$

and for the alkaline-earth (bivalent) metals it is

$$\text{Ba}^{+2} > \text{Sr}^{+2} > \text{Ca}^{+2} > \text{Mg}^{+2},$$

at least in resins of the strong-acid type.

7.123. It is important to note that it is the radius of the hydrated ion, i.e., of the ion as it exists in solution, that determines the extent to which the particular ion will exchange with a resin. This is usually quite different from the radius of the bare ion, i.e., the ion as present in a solid compound, generally referred to as the crystal radius. In both the alkali metal and alkaline-earth metal series the order of increasing size of the hydrated ions is the reverse of that of the crystal ions.

7.124. In the rare-earth (lanthanide) series of elements the radii of the hydrated tripositive ions increase regularly with increasing atomic number. Hence the affinities of the rare-earth ions for a particular resin decrease as the atomic number increases. The same general rule is found to apply to the tripositive ions of the actinide series, so that the retention decreases in the order

$$U^{+3} > Np^{+3} > Pu^{+3} > Am^{+3} > Cm^{+3}.$$

7.125. Although the retention of anions is generally determined by the same conditions as mentioned above for cations, the behavior is not so well defined. The order of the affinities of a strong-base resin for a number of common anions is approximately

$$SO_4^{-2} > CrO_4^{-2} > NO_3^- > HPO_4^{-2} > I^- = Br^- > Cl^-.$$

The apparently incorrect position of phosphate ions may perhaps be accounted for by the relatively large size of these ions, which outweighs the effect of their charge.

Separations by Ion Exchange

7.126. The varying affinities of different ions for a given resin can be used as the basis of a technique for the separation of such ions from each other. The procedure is carried out in two stages, namely, (1) sorption and (2) elution. In the sorption step the solution containing the ions to be separated is poured into the top of a column (or bed) filled with fine porous grains of the resin in an appropriate form, e.g., H—R for cations or R—Cl (or R—OH) for anions. The ions in solution undergo exchange with the mobile ions of the same sign on the resin, so that the former are gradually taken up (or sorbed) completely as the solution proceeds down the column.

7.127. The elution stage represents the actual separation step. A suitable solution, called the eluting solution or the *eluent*,* is allowed to percolate slowly through the ion-exchange column containing the sorbed ions. These ions are continuously removed and redeposited as the eluent flows down the column. If the eluent has been properly chosen there is a degree of selectivity in these processes, so that the *eluate*, i.e., the liquid emerging from the bottom of the column, will contain first one, and then another, of the ions in a relatively pure form.

7.128. The conditions for good separation may be understood, in a general way, from the following considerations. The affinity (or retention) of a particular ionic species for a given resin may be expressed by the distribution ratio (or coefficient), K_d. This is defined as the ratio of the concentration of the ion in the resin to that in the solution at equilibrium, i.e.,

$$K_d = \frac{\text{Concentration of ion in resin}}{\text{Concentration of ion in solution}}$$

at equilibrium. For two species, A and B, the *separation factor*, α, for a given resin, is the ratio of the distribution coefficients for that resin; thus,

* The terms eluant, elutrient, and elutriant are to be found in the literature, but to the present writer they seem less appropriate than eluent.

$$\alpha \equiv \frac{K_d(\text{B})}{K_d(\text{A})} = \frac{\text{Conc. of B in resin}}{\text{Conc. of A in resin}} \times \frac{\text{Conc. of A ions in solution}}{\text{Conc. of B ions in solution}}$$

or

$$\frac{\text{Conc. of B in resin}}{\text{Conc. of A in resin}} = \alpha \times \frac{\text{Conc. of B ions in solution}}{\text{Conc. of A ions in solution}}. \qquad (7.128.1)$$

7.129. In order to achieve good separation of A and B in the elution stage it is necessary that the left side of equation (7.128.1) be made as large as possible. The species B will tend to be retained by the resin while A is removed by the eluent; A ions will thus move steadily ahead of B in the ion-exchange column during elution and will leave the column first.

7.130. It will be seen from equation (7.128.1) that the ratio of B to A remaining on the resin can be made large in two ways: first, by having α large and the ratio of the ionic concentrations of B to A in solution not too small, and, second, if α is neither large nor small, e.g., about unity, the ratio of the B ions to A ions in solution must be large. These two conditions will be examined in turn.

7.131. In view of the definition of the separation factor given above, a large value of α means that $K_d(\text{B})$ must be greater than $K_d(\text{A})$; in other words, B must have a larger affinity for the resin than does A. In this event satisfactory separation can be achieved when the ions present in the solution are at more or less equivalent concentrations. These conditions can be realized, as a general rule, when A and B represent ions carrying different charges; for example, A might be Na^+ and B could be Mg^{+2}, so that the latter has a greater affinity for a cationic resin than does the former. If these ions were sorbed on a resin in the

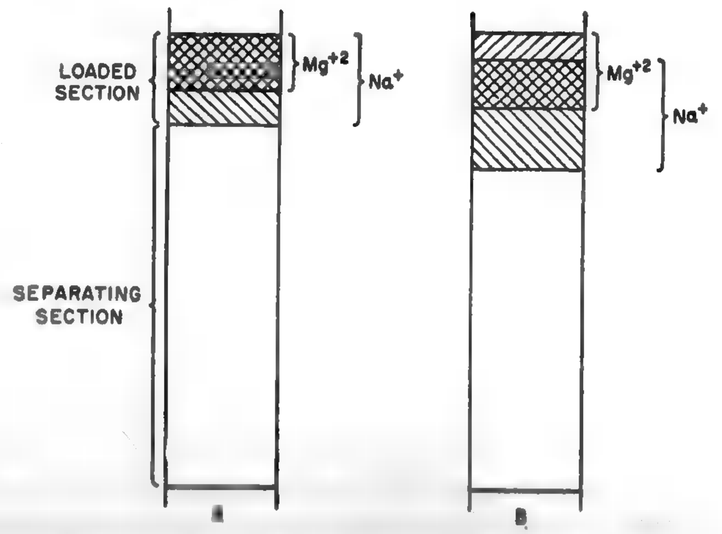


Fig. 7.7. A. Ion-exchange column before elution
B. Ion-exchange column during elution

H—R form and then eluted with a dilute solution of a simple acid, e.g., hydrochloric acid, the Na^+ ions would move steadily ahead of the Mg^{+2} ions, as indicated in Figs. 7.7 A and 7.7, B. The first portion of the eluate to leave the column will consist almost entirely of dilute acid and this will be followed by eluate containing Na^+ ions with very little or no Mg^{+2} ions. The final eluate will contain the Mg^{+2} ions only. The same general behavior would be observed for a solution containing three or more ions, provided they had appreciably different affinities for the resin.

7.132. The procedure just described could also be used to separate anions having markedly different ion-exchange affinities, e.g., SO_4^{-2} and Cl^- ions. The anionic resin would be used in the hydroxylic form, i.e., R—OH, and after sorption the ions would be eluted with a dilute solution of an alkali, such as sodium hydroxide for a strong-base resin or ammonia for a weak-base type. In the case mentioned, the eluate would contain first the Cl^- ions and then the SO_4^{-2} ions.

7.133. When the affinities of the ions for a given resin are not very different the foregoing separation technique will not be efficient. This will be the case, for example, when it is required to separate a number of ions having the same charge, e.g., the tripositive ions of the lanthanide or actinide series. It is then necessary to make use of the second condition mentioned in § 7.130, namely, to make the ratio of B ions to A ions in solution as large as possible. This can be achieved by employing an eluting solution containing anions which form complexes with A and B of differing stabilities. For example, if the complex formed with A is stronger than that formed with B, the ratio of free B ions to free A ions in solution will be large and the conditions for satisfactory separation of A from B will be realized. The earlier portions of the eluate will contain almost pure A and the later portions pure B.

7.134. For the separation of tripositive ions, such as those of the lanthanide and actinide series, the most widely used eluent has been a solution of citric acid partially neutralized with ammonia to a pH of about 3 to 3.5. This solution contains the H_2Cit^- anion which forms neutral complexes, having the general formula $M(H_2Cit)_3$, with tripositive ions. The stability of the complexes in each series increases with decreasing atomic radius, i.e., with increasing atomic number. Hence upon elution with the citric acid solution mentioned, the elements appear in order of decreasing atomic number, i.e., in order of decreasing stability of the complexes in solution.

7.135. The two separation procedures considered may be regarded as extreme cases and various combinations are possible. The availability of both cationic and anionic resins and the ability to form complexes of various types make possible a considerable variety of separation methods. For example, although the primary application of anion-exchange resins is for the separation of anions, they can also be employed to separate some metallic elements, which normally form cations in sdution. Use is made of the fact that, in the presence of excess

of certain anions in solution, many elements, as already seen (§ 7.35), form negative, i.e., anionic, complex ions. These can then exchange with the anions of a resin.*

7.136. As an illustration, consider the separation of ferric iron (Fe^{+3}) and potassium (K^+) ions. If the solution contains traces of Fe^{+3} ions in the presence of a large proportion of K^+ ions, the most practical method of separation is to add an excess of chloride to the solution. The iron forms negatively charged complexes and so can be removed from solution by an anion-exchanger resin, but the potassium, which does not complex, passes through the column. The iron held by the resin can subsequently be removed by elution with a dilute chloride solution, e.g., dilute hydrochloric acid. Since the (negative) complex ferric anions are less stable at low concentrations of chloride, they are removed from the resin and are replaced by chloride ions.

7.137. On the other hand, if the potassium were present in trace amounts and the ferric ions were in excess, the latter could be converted into an anionic complex by chloride ions and the solution passed through a cation-exchange resin. The potassium would undergo exchange with the hydrogen of the resin and be retained, but the complex ferric anions would pass through. The potassium could be removed from the resin by elution with an acid solution.

7.138. Anion-exchange resins are particularly valuable for separating highly charged cations, which are relatively easily hydrolyzed. In the fairly dilute acid solutions frequently used for sorption by a cation-exchange resin, such ions would be partially, at least, in the colloidal state. In this form there would be no exchange with the resin, and the material would pass through the resin bed. The difficulty due to colloid formation can often be overcome by converting the metallic ions into complex anions and employing anion-exchange resins.

General Ion-exchange Applications

7.139. Of the four applications of ion-exchange resins referred to in § 7.119, the basic principles of ion separation have been discussed. Before proceeding to consider other applications, some aspects of the ion-exchange procedure, which are of general significance, must be mentioned. Since the resins have a limited capacity for sorbing ions they are not suitable for removal of ions present at high concentrations. Nevertheless, an ion of low concentration may often be separated from one of high concentration by the use of some such procedure as is described in § 7.136 and § 7.137. Another matter to be borne in mind in connection with the use of ion exchange is the affinities of the ions for a given resin. If there is no appreciable complex formation, the ion with the greater affinity will tend to displace the one with lower affinity from the resin. But the situation can be greatly affected by the presence, either in the original solution or in the eluent, of complex-forming ions.

* It should be recalled, in this connection, that the citrate complexes formed in citric acid solution of pH about 3 are neutral and so do not participate directly in ion exchange.

7.140. If it is required to remove an undesired ion from solution, the simplest procedure is to pass the solution through a resin which has a strong affinity for that ion. It is important to remember, however, that the undesired ion will be replaced in solution by the one of the same sign originally attached to the resin. If removal of the unwanted ion is to be effective, it must be able to displace the ion already on the resin. A common procedure for the removal of small amounts of dissolved salts from water is to pass the water first through a cationic resin in the H—R form so that the cations present are replaced by hydrogen ions. The water is then allowed to percolate through an anionic resin in the R—OH condition, so that the anions are replaced by hydroxyl ions. In some cases the two resins may be mixed in a single (mono-bed) column. Thus the cations and anions in the water are removed and their place is taken by hydrogen and hydroxyl ions which immediately combine to form water.

7.141. When a wanted ion is to be extracted from a solution, the latter is passed through an ion-exchange bed which will take up the ion. The sorbed material must now be removed from the resin by means of a suitable eluent. The presence of a complex-forming substance in this solution will greatly facilitate the removal. The same principle can be used to concentrate a particular ion originally present in a dilute solution. It is sorbed on a resin and then extracted with an effective eluent.

7.142. The method of causing metathesis, or replacement of one ion by another, by means of an ion-exchange resin is obvious, although the exact procedure may vary according to the circumstances. If the ion already in solution has the stronger affinity for a resin, then the ion to be exchanged for it is first sorbed on the resin. It is then eluted with the given solution so that the required interchange occurs. If the reverse is the case, so that the ion initially in the solution has the weaker affinity, the solution is passed through an ion-exchange resin so that the ion in question is sorbed. The resin is then eluted with a solution containing the other ion.

7.143. In connection with the operation of nuclear reactors there are various directions in which ion-exchange procedures might find some application. The treatment of solutions obtained from spent reactor fuel presents some difficulty because of the large proportion of uranium and the very small amounts of plutonium and fission products. A possible method of separation would be to pass the solution through a resin which sorbed the plutonium and fission products but not the uranium. The plutonium could then perhaps be eluted in a suitable manner. Another possibility would be to find such conditions that only the plutonium was sorbed on an ion-exchange resin, the uranium and fission products passing out with the effluent.

7.144. The ion-exchange technique is widely employed in water treatment, and a similar process could be used to remove corrosion products and radioactive ions formed as a result of neutron capture in reactor cooling water. The same method could be adapted to the purification of water for various chemical

processing operations. Ion exchange might also be used for the separation, purification, and concentration of individual fission products of value for experimental or industrial purposes. As will be seen later, one of the problems of reactor fuel processing is the disposal of the dilute solutions of radioactive waste material. It is possible that concentration by ion exchange will alleviate the situation to some extent. However, a difficulty that may arise here, as with all operations involving radioactive solutions, is the instability of the organic resins to beta and gamma radiation.

Ion-exchange Column Design

7.145. As the eluent moves down a column of ion-exchange resin the adsorbed ions are repeatedly removed from and taken up again by the resin. The process is analogous to that occurring in a distilling column, where the vapor may be regarded as successively condensed and vaporized. The difference in the affinities of the ions (or the tendency to form complexes) results in a partial separation of the ions at each stage and the effect is multiplied many times over the length of the column. If the ionic concentration of the eluate is determined at various elution times, the results will be somewhat as shown in Fig. 7.8, based on the supposition that the A ions are eluted first. The concentration of A ions in the eluate increases to a maximum and then decreases; this is followed by a similar increase and decrease in the B ion concentration. It will be seen that there is an intermediate region in which the eluate contains both ions. The extent of this region is smaller the greater the effective separation factor,* α, and the longer the length of the column in which fractionation occurs. This behavior is again similar to that observed in the fractional distillation of a mixture of two liquids. The first and last fractions are essentially pure, but the fraction in between contains both components. The volume of this intermediate fraction is less the greater the difference in the vapor pressures of the liquid and the longer the distilling column.

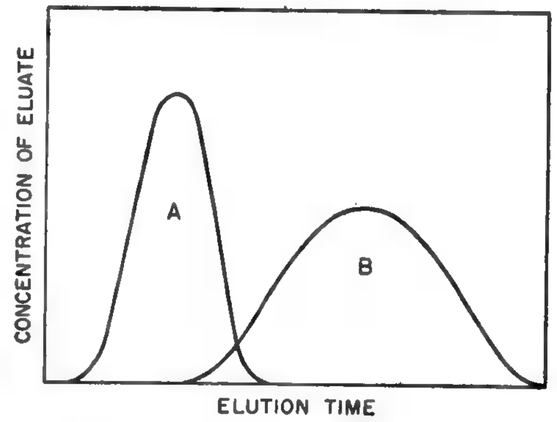


Fig. 7.8. Elution curves for ions A and B

7.146. The efficiency of a given ion-exchange column, under specified conditions, can be expressed in terms of the equivalent number of theoretical plates (or stages) as in distillation in which unsteady state conditions are operative.

* The term "effective separation factor" is used here in the sense that the effect of complexion formation is included.

Thus a theoretical stage may be defined as any portion of the column, e.g., $AABB$ in Fig. 7.9, such that the eluent leaving the lower end (BB) will have the same composition as a solution in equilibrium with the ion-exchange resin at the upper end (AA). If equilibrium between the eluting solution and the resin were attained instantaneously, it is probable that for particles of finite size the theoretical stage height would be roughly the same as the diameter of the resin particles. In reality, however, the attainment of equilibrium involves one or more slow steps (usually diffusion across a boundary layer) and the actual height of a theoretical stage may range from a few millimeters to several centimeters.*

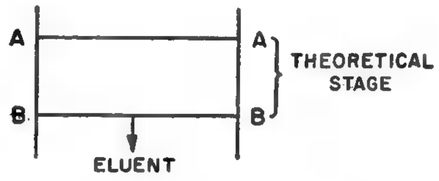


FIG. 7.9. Representation of theoretical stage for ion exchange

7.147. The closer the approach to equilibrium between the ions in the eluting solution and those attached to the resin, the greater the number of theoretical stages per unit length of column and the more efficient will the given column be for ion exchange. The conditions which favor the rapid attainment of equilibrium are (1) small resin particles of high porosity, (2) slow rate of flow of the eluent through the column, (3) high temperature, and (4) low ratios of cation to resin, i.e., dilute solutions. All these tend to increase the number of theoretical stages in a given column and, hence, the efficiency of ion separation for a particular value of the theoretical separation factor.

7.148. From the practical point of view, some of the foregoing conditions are undesirable. For example, if the resin particles are too small, the pressure drop through the column will be too great; slow flow rates and low cation-to-resin ratios mean low yields per unit time. The number of theoretical stages can be increased, of course, by increasing the length of the resin column, but this means that larger volumes of eluent have to be used and that the elution time is increased. The actual conditions employed in any particular case must thus represent a balance between the separation efficiency of the column, on the one hand, and the desire for high yields, on the other.

7.149. The length of column required to produce a required efficiency of separation can be calculated, provided the equilibrium values of the (effective) distribution coefficient of the ions in the given eluent are known and an experimental elution curve has been determined in a column of the same resin. This curve is generally of the form shown in Fig. 7.10, in which the ordinate is the relative concentration of the particular ion in the eluate and the abscissa is F, the number of so-called "free volumes" of eluate that have been collected. The free volume is the liquid phase volume, i.e., the volume of the resin column minus the actual volume of the resin particles. The number of free volumes of solution that have come from the column when the concentration of the ions in the eluate is a maximum is represented by C. It can be shown theoretically that C is equal to the ratio of the fraction of the given ion in the solution to the fraction in the resin. The value of C can be calculated from the distribution coefficient, thus permitting a check on Fig. 7.10.

* This height may vary with the position in the column, especially if the effective separation factor of the ions is not constant.

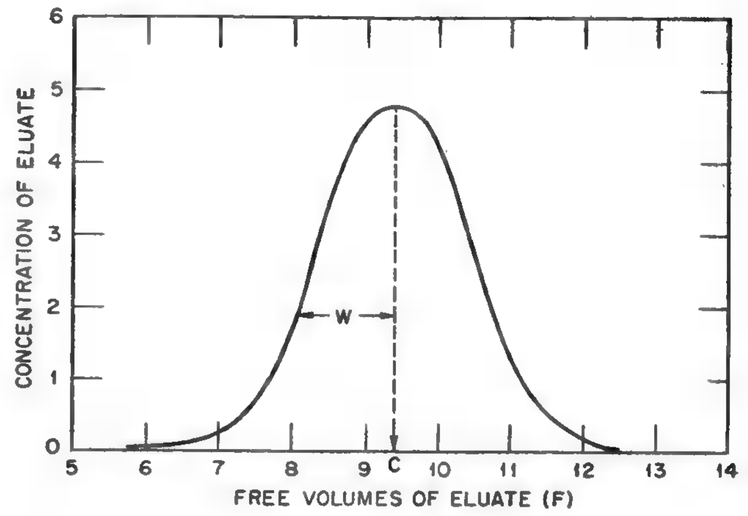


FIG. 7.10. Calculation of number of theoretical stages from elution curve (S. W. Mayer and E. R. Tompkins)

7.150. The number of theoretical stages, n, in the column used can be expressed approximately by

$$n = \frac{2C(C+1)}{W^2}, \tag{7.150.1}$$

where W is the half width of the elution curve at an ordinate value that is $1/e$ of the maximum, as shown in Fig. 7.10, e being the base of natural logarithms. In the particular case illustrated C is 9.41 and W is 1.38; hence,

$$n = \frac{(2)(9.41)(10.41)}{(1.38)^2} \approx 103,$$

so that for the given experimental conditions the column is equivalent to about 103 theoretical stages.

7.151. Since n is now known and C may be regarded as available from the distribution coefficient for any ion, other than the one used to obtain Fig. 7.10, the value of W for that ion, in the existing circumstances, can be calculated from equation (7.150.1). By assuming that the elution curve has the form of a normal (Gauss) error curve, an assumption which becomes increasingly valid as the number of theoretical stages increases, it should now be possible to construct the elution curve for any ion. By placing the curves for different ions

together, the extent of overlap, and hence the effectiveness of the separation, can be determined.

7.152. In actual practice the procedure would be reversed. The amount of overlap which could be tolerated would be prescribed and the number of theoretical stages required to produce this degree of separation would be calculated. The number of theoretical stages in a column is approximately proportional to its height, and so the height of the required column can be estimated.*

7.153. The design calculations outlined above are based on the use of a fixed bed of resin through which the various solutions percolate. Such fixed beds have hitherto been in general use both in industry and in the laboratory; they represent one of the main disadvantages of the ion-exchange technique, since operations must be carried out on a batch, rather than on a continuous, basis. This problem is, however, being given serious consideration. Experimental models of moving-bed columns have already been designed, and practical forms suitable for large-scale operation will undoubtedly be developed in due course.†

SEPARATIONS BY SOLVENT EXTRACTION ‡

General Principles of Solvent Extraction

7.154. The solvent-extraction method for separating components of an aqueous solution can be used when one of the components is not only appreciably soluble, but is more soluble than the other components, in an organic liquid which is essentially immiscible with water. When the organic solvent is intimately contacted with the aqueous solution, the substances present will distribute themselves between the organic and aqueous media in a more or less definite manner, depending on their relative solubilities in the two phases. The component (or components) with the greatest solubility in the organic solvent will tend to pass into that phase. Thus a partial separation of the components of the aqueous solution will have been achieved. The degree of separation can be enhanced by back-extracting the substances in the organic phase into an aqueous solution and then re-extracting with fresh organic solvent.

7.155. As an engineering problem, solvent extraction is to be regarded as a diffusion operation involving the transfer of a solute from one liquid phase to another liquid phase with which it is essentially immiscible. By means of suitable columns (or other devices) using the countercurrent principle, with one liquid flowing in one direction and the other liquid in the opposite direction, a

* For a description of the basic theory and some applications, see S. W. Mayer and E. R. Tompkins, *J. Am. Chem. Soc.*, **69**, 2866 (1947); also, E. R. Tompkins, *J. Chem. Ed.*, **26**, 32, 92 (1949), especially p. 96, *et seq.*

† Cf. I. R. Higgins and J. T. Roberts, Report AECU-2546; S. H. Jury, ORNL Report CF-53-1-213; *Nuc. Sci. Abs.*, **8**, Abs. No. 173 (1954); N. K. Hiester, *et al.*, Report AECU-2742 (Final Summary Report); *Chem. Eng. Prog.*, **50**, 139 (1954).

‡ For general references, see T. K. Sherwood and R. L. Pigford, "Absorption and Extraction," McGraw-Hill Book Co., Inc., 1952; R. E. Treybal, "Solvent Extraction," McGraw-Hill Book Co., Inc., 1951; J. H. Perry (Ed.), "Chemical Engineers' Handbook," 3rd ed., McGraw-Hill Book Co., Inc., 1950, pp. 714–753.

continuous, steady-state operation can be achieved. Because no solid phases are involved, liquid-liquid extraction is perhaps one of the simplest unit processes that can be operated by remote control; this is a matter of great significance in connection with the treatment of highly radioactive materials, such as spent fuel elements or fission products alone.

7.156. As compared with ion exchange, solvent extraction has the disadvantage of smaller mass-transfer efficiency, i.e., the height equivalent to a theoretical stage is greater. On the other hand, the separation factors are frequently larger for solvent extraction. In addition to the possibility of continuous operation by remote control, the solvent-extraction procedure has the advantage of being applicable to concentrated solutions, whereas ion exchange is largely restricted to dilute solutions.

Choice of Organic Solvent

7.157. In choosing the organic liquid to be used for a particular solvent-extraction procedure, various factors must be taken into consideration. These usually represent independent properties of the solvent, and the actual substance chosen may involve a compromise giving the best combination of desirable characteristics. The most important property is the selectivity or ability of the organic liquid to extract a particular component (or components) of a solution in preference to all others that are present. The selectivity is expressed by the separation factor which, as in § 7.128, is the ratio of the distribution coefficients of the wanted and unwanted species when equilibrium is attained between the two phases. The distribution coefficient or distribution ratio, D, is defined as

$$D = \frac{\text{Conc. of component in organic phase}}{\text{Conc. of component in aqueous phase}} \qquad (7.157.1)$$

at equilibrium, and the separation factor, α, is given by

$$\alpha = \frac{D(\text{product})}{D(\text{impurity})}.$$

A good solvent for extraction is one for which the distribution coefficient for one component is large and the separation factor is either large or small. In other words, it is desirable that D(product) shall be large, whereas D(impurity) should be small, or vice versa.

7.158. Ideally, the organic solvent should be immiscible, or almost immiscible, with the solution to be extracted. In practice complete immiscibility is not possible since all liquids exhibit some mutual solubility. However, in some cases this solubility is less than in others, and the solvent chosen should be slightly soluble (2 per cent at most) in water.

7.159. The organic liquid should have sufficient chemical stability so that it does not interact to any appreciable extent with the various chemical reagents used in the extraction process. It may have to withstand the action of fairly high concentrations of oxidizing and reducing agents. In addition, the organic

solvent must be stable to the intense beta and gamma radiations emitted by the fission products as well as to the alpha particles from uranium and plutonium.

7.160. In order that the two liquid phases may separate readily following contact, it is necessary that the density of the organic solvent should differ appreciably from that of the aqueous solution being extracted. The difference in the densities of the two phases will largely determine the nature and size of the extraction equipment. The greater the difference the more rapidly will the phases tend to separate and, hence, the smaller will be the cross-sectional area of the contacting equipment for a given product throughput rate.

7.161. The interfacial tension between the organic liquid and the aqueous medium should be high enough to prevent the formation of a stable emulsion since this would hinder separation of the two phases. On the other hand, the interfacial tension must be low enough to permit good dispersion of one liquid phase in the other, thus ensuring a large area of contact between them. The viscosity of the organic solvent must be relatively low to facilitate flow through the contactor and to minimize pumping requirements.

7.162. Although the foregoing properties of the solvent may be regarded as essential, there are certain others which are, at least, highly desirable. It should be readily available in a pure state at a reasonable cost, and it should be easily recoverable, e.g., by distillation. From the standpoint of industrial safety, it is desirable that the solvent have both low toxicity and low inflammability.

7.163. The simplest inorganic compounds which have lent themselves to solvent extraction are the nitrates, although some work has also been done with chlorides. Many complexes formed by metals with organic molecules, known as "chelate" compounds, can be readily extracted from aqueous solutions. In this connection the complex compounds formed with α-thenoyl trifluoracetone have been found useful for the separation of cations.*

7.164. For the extraction of nitrates of high-valence ions, such as uranium (VI), thorium (IV), and cerium (IV) a wide variety of organic liquids have been used, particularly ethers, ketones, alcohols, and esters.† Mention may be made, in particular, of diethyl ether, methyl isobutyl ketone, amyl alcohol, and amyl acetate. An organic solvent which has been found to be particularly resistant to oxidizing agents is *n*-tributyl phosphate.‡ Because the density of this liquid is close to that of water and its viscosity is high, it may be necessary to add a diluent, e.g., carbon tetrachloride or butyl ether.

Factors Influencing Solvent Extraction

7.165. The extraction of an inorganic compound, such as a nitrate, from aqueous solution by means of an organic solvent is affected by a number of circumstances. Three factors of particular importance will be considered here; they are the presence of (1) salting agents, (2) oxidizing or reducing agents, (3) complex-forming anions.

* See, for example, M. Calvin and J. C. Reid, *J. Am. Chem. Soc.*, **72**, 2948 (1950).

† L. I. Katzin and J. C. Sullivan, Report AECD-2537; see also C. C. Templeton and N. F. Hall, *J. Phys. Chem.*, **51**, 1441 (1949); H. M. Irving, *Quarterly Rev.* (Chem. Soc. London), **5**, 200 (1951).

‡ J. C. Warf, *J. Am. Chem. Soc.*, **71**, 3257 (1949).

7.166. A *salting agent* is either a salt or an acid the anion of which is the same as that of the inorganic compound to be extracted. For example, for the extraction of a nitrate, such as uranyl (VI) nitrate, either nitric acid or a neutral nitrate, such as sodium, potassium, or calcium nitrate, which is soluble in water but not in the organic liquid, might act as salting agent. The effect of a salting agent in the aqueous solution is to increase the distribution coefficient as defined by equation (7.157.1). In other words, the concentration of the inorganic

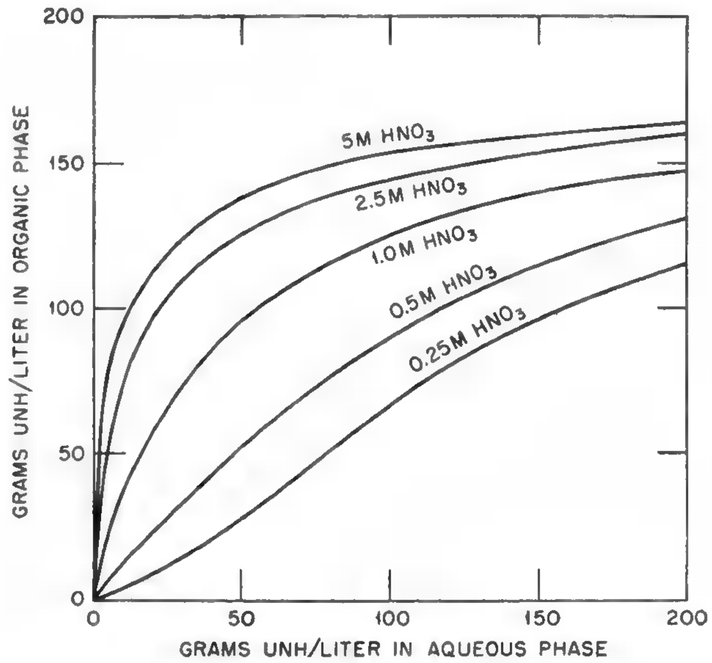


Fig. 7.11. Distribution of uranyl nitrate hexahydrate between organic and aqueous phases (R. L. Moore)

compound under consideration in the organic phase is increased relative to the concentration in the aqueous phase when a salting agent is present. This can be seen from Fig. 7.11, in which the ordinates are the concentrations of uranyl nitrate hexahydrate (UNH) in an organic medium, consisting of a mixture of tributyl phosphate (20 per cent) and carbon tetrachloride, and the abscissae are the corresponding equilibrium concentrations in aqueous solutions containing the indicated amounts of nitric acid as salting agent.*

7.167. It is evident that in the absence of nitric acid the uranyl nitrate has a preference for the aqueous phase, but when the latter contains 5 M nitric acid, the uranyl compound, especially at low concentrations, preferentially enters the organic phase. Extraction of uranyl nitrate by the organic solvent would thus

* R. L. Moore, Report AECD-3196.

be greatly favored by the presence of appreciable quantities of nitric acid in the aqueous solution. A large proportion of the uranium could then be back-extracted into the aqueous phase, if required, by bringing the organic solution into contact with water containing little or no nitric acid.

7.168. According to the statement in § 7.157, efficient separation of one substance from another by solvent extraction is favored by a high value of the distribution coefficient for the desired product and a low value for the undesired (impurity) substances. If the salting agent affected both distribution coefficients to the same relative extent, the separation factor would remain unaltered and there would be no advantage from the standpoint of extraction. The condition to be aimed at is to increase the distribution coefficient of the desired product, by the addition of a suitable salting agent, but to leave the values for the impurities essentially unchanged.

7.169. The distribution coefficient of the nitrate of a particular element is markedly affected by the oxidation (or valence) state of that element. It is for this reason that the presence of oxidizing or reducing agents in the aqueous solution can completely change the extractability by an organic solvent. In general, the nitrates of the actinide elements in the (VI) state can be extracted in this manner, but they are much less extractable in the lower oxidation states. To judge from the fact that the distribution coefficients of the tripositive nitrates of the rare-earth (lanthanide) elements between organic media and water are very low, it is to be expected that the nitrates of the actinide elements in the (III) state will not be easily extracted by organic solvents.

7.170. One of the consequences of the influence of the oxidation state on the distribution coefficient is that after an element has been extracted into an organic medium it can often be back-extracted into aqueous solution by adding a reducing agent to the latter. If two actinide elements, e.g., uranium and plutonium, have been extracted in the (VI) state into an organic solvent, this fact can be used as a basis for separating these elements. The separation can be achieved by back-extracting the organic phase with an aqueous solution which will reduce one of the actinide elements but not the other. As seen earlier in this chapter, plutonium (VI) can be reduced much more readily than uranium (VI), and so the former could be back-extracted with a suitable reducing agent leaving the uranium in the organic phase.

7.171. The extraction of a specified element from aqueous solution by an organic medium is dependent upon the particular form in which the element is present in the solution. For example, uranyl nitrate hexahydrate can be extracted by diethyl ether, but the corresponding sulfate is not extractable by this solvent. The addition of a sulfate or other salt of a complex-forming anion to an aqueous solution of uranyl nitrate will thus decrease the extractability of the uranium, since a proportion of the element will be in some form other than the nitrate. The complex-forming anions decrease the distribution coefficient between the organic and aqueous phases, and so their effect is opposite to that of the common-ion salting agents referred to in § 7.166.

7.172. Another aspect of complex formation is its effect on the oxidation-reduction potential, mentioned earlier. Accordingly salts of complex-forming anions may be able to influence the extraction (or back-extraction) of a given element in the presence of a specified reducing or oxidizing agent. It is apparent, therefore, that the extractability of an element can be affected by variations in the amount and nature of the salting agent, by the nature of an oxidizing or reducing agent, and by the presence of complex-forming ions. The possibility of changes in these three factors thus permits flexibility in the application of the solvent-extraction method for the separation of various elements.

Extraction, Strip, and Scrub Columns

7.173. Solvent extraction is usually carried out in some form of contactor (§ 7.188), using the countercurrent principle, as stated earlier. It is the purpose of the present section to describe briefly the general procedure adopted in the performance of a solvent-extraction cycle. For simplicity, it will be assumed that the down-flowing phase I is the aqueous solution from which the desired solute is to be extracted by the up-flowing organic solvent phase II. The flow paths for countercurrent extraction would then be as represented in Fig. 7.12. The entering aqueous solution is called the *feed*, and that leaving the column after extraction is sometimes referred to as the *raffinate*. The organic solution containing the extracted substance is known as the *extract*. If the density of the organic solvent is greater than that of the aqueous solution, the directions of flow would be reversed. The feed would enter at the bottom and the solvent at the top, whereas the raffinate would leave at the top and the extract at the bottom.

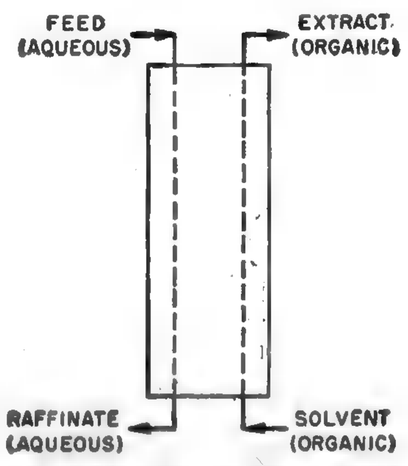


Fig. 7.12. Simple liquid-liquid extraction column

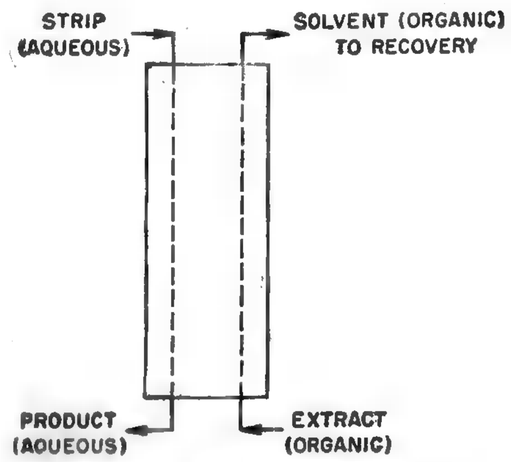


Fig. 7.13. Simple strip column for back-extraction

7.174. After the desired solute has been extracted into the organic phase, it is usually necessary to back-extract it into an aqueous solution. This process, called *stripping*, is carried out as indicated in Fig. 7.13. The theoretical treat-

ment of a strip column is exactly analogous to that of an extraction column except that phase I may now be regarded as the organic phase and phase II as the aqueous phase (see § 7.182, *et seq.*).

7.175. As a general rule the organic extract contains, in addition to the desired substance, e.g., uranium or plutonium, small amounts of undesired substances, e.g., fission products. These may be removed, partially at least, by passing the organic solution through a *scrub* column, i.e., a form of strip column in which the less soluble part of the dissolved material, e.g., fission products, is back-extracted into the aqueous solution. A simpler scheme is depicted in Fig. 7.14, which is, essentially, a combination of Figs. 7.12 and 7.13. The lower part of the column is the extraction section, and the upper is the scrub section. The aqueous feed solution enters the column at the middle and flows downward. The organic solvent flowing upward extracts the desired solute as well as some of the more easily extracted impurities. In the upper portion of the column, most of the latter are removed by scrubbing the organic phase with a suitable aqueous solution. The organic extract leaving the top of the column is thus relatively pure. The scrub solution will, of course, also take up some of the desired solute, but this is re-extracted into the organic phase as it flows down in the lower section of the column. The general behavior of the extraction system is similar to that of a reflux column in distillation.

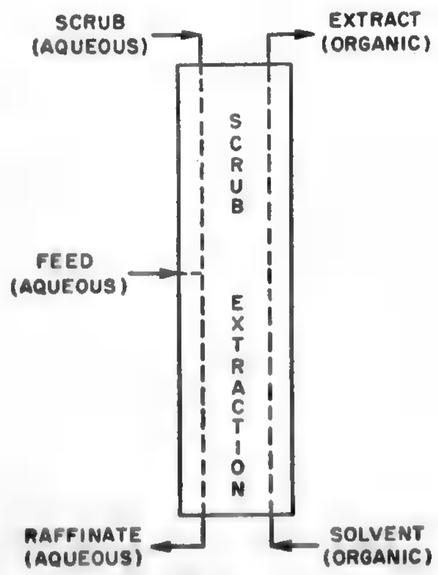


Fig. 7.14. Extraction column with scrub section

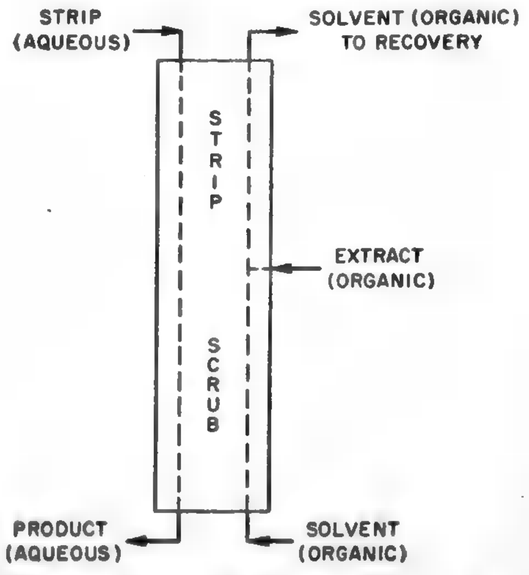


Fig. 7.15. Back-extraction (strip) column with scrub section

7.176. A scrub section is frequently attached to a strip column when it is desired to back-extract only one of two or more components of an organic extract. An example is the stripping of plutonium from an organic phase containing both plutonium and uranium as described below. In a case of this kind the upper part of the reflux column constitutes the strip section, while the lower is the scrub section (Fig. 7.15). The organic feed solution, entering at the middle of the column, flows upward and is stripped by the aqueous stripping solution. In the lower section of the column this is scrubbed with fresh organic solvent, entering at the bottom, which re-extracts the unwanted solute. The stripped product, e.g., plutonium, leaves in the aqueous solution from the bottom of the column, while the other solute (or solutes), e.g., uranium, is in the organic phase leaving the top of the column.

Applications of Solvent Extraction

7.177. The application of the basic principles of solvent extraction, described above, to the separation of uranium and plutonium from each other and from the fission products present in spent reactor fuel may be illustrated by means of Fig. 7.16.* The fuel elements are dissolved in nitric acid and then oxidized so that the uranium and plutonium are present as nitrates of the (VI) state, i.e., $UO_2(NO_3)_2$ and $PuO_2(NO_3)_2$, respectively. The fission product nitrates are represented by $F.P.(NO_3)_x$.

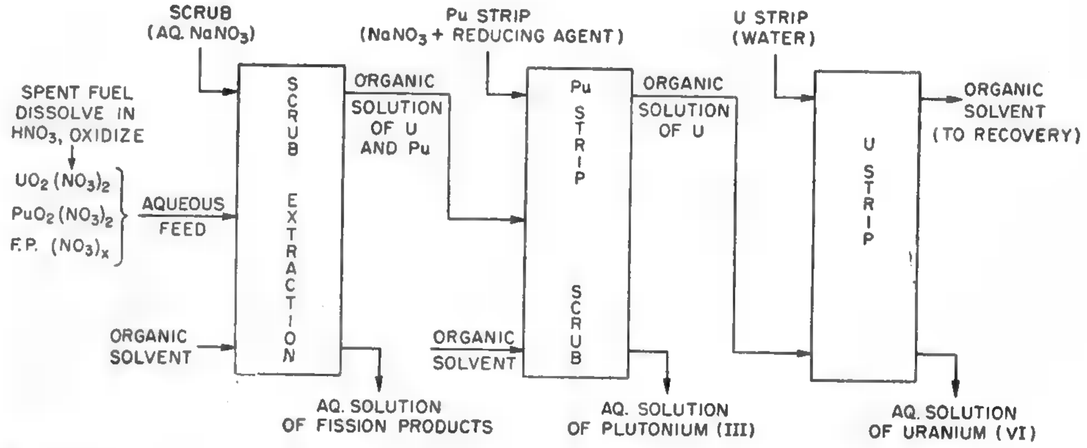


Fig. 7.16. Separation of plutonium and uranium from spent fuel by solvent extraction

7.178. The aqueous solution containing the uranium, plutonium, and fission product nitrates, which is the feed solution, enters at the middle of the first column and flows downward, while a less dense organic solvent, entering from the bottom, flows upward. A considerable proportion of the uranium and plutonium thus passes from the aqueous to the organic phase. In the upper (scrub) part of the column the organic phase is scrubbed with an aqueous solution of sodium nitrate which acts as a salting agent. Any fission products that have been extracted by the organic solvent are now back-extracted into the

* F. R. Bruce, Report AECD-3449; J. F. Flagg and E. L. Zebroski, *loc. cit.*

aqueous phase, but the salting agent largely prevents back-extraction of the uranium and plutonium. The aqueous effluent (raffinate) from the extraction column contains essentially all the fission products with little or no uranium or plutonium.

7.179. The organic phase containing the uranium and plutonium next passes into the second (stripping) column where it flows upward and meets the down-flowing aqueous strip solution containing a suitable reducing agent as well as the salting agent, sodium nitrate. The plutonium, in a lower oxidation stage, e.g., plutonium (III), is back-extracted into the aqueous phase and as this flows downward it is scrubbed with fresh organic solvent flowing upward from the bottom of the column. Any uranium (VI) that has passed into the aqueous solution is thereby returned to the organic phase. The aqueous medium, containing plutonium (III) nitrate, leaves the column at the bottom.

7.180. The organic solution of uranium (VI) nitrate, from which the plutonium and fission products have been almost completely separated, is now transferred to the bottom of the third (strip) column where it flows upward and is stripped by water flowing downward. In the absence of a salting agent the uranium is back-extracted into the aqueous phase and flows out of the bottom of the column. The used solvent, leaving at the top, is sent to a recovery plant for purification and subsequent re-use in extraction.

Design of Extraction Columns

7.181. The design of columns for liquid-liquid extraction is a well-developed field of chemical engineering.* The primary objective of such design is to effect the maximum transfer of one or more dissolved substances from one liquid phase to another. The calculations are based on the use of steady-state diffusion theory. For a complete treatment it is necessary to study the equilibrium distribution between the two phases of all the chemical constituents of the system over the whole range of concentrations that may be encountered, i.e., from the concentration of the feed solution to that of the aqueous waste. From these data can then be determined the optimum flow rates of the two liquids that will minimize both the number of theoretical stages of extraction and the volume of the organic extractant required.

7.182. The height equivalent to a theoretical stage for a specified degree of extraction can be estimated by the methods given in standard texts. Consequently this topic will not be considered here, although it is of interest to examine the relationship between the number of stages and the relative flow rates of the liquid phase. If n is the number of theoretical stages required to decrease the concentration of a given solute from x_0 to x_n, in a particular phase, then it can be shown from material balance considerations that, provided the distribution coefficient is independent of the solute concentration and the two liquid phases are essentially immiscible,

* See references to § 7.154.

$$\frac{x_n}{x_0} = \frac{1}{R^n + R^{n-1} + R^{n-2} + \cdots + 1} = \frac{R-1}{R^{n+1}-1}, \tag{7.182.1}$$

where R, called the *extraction factor*, is defined by

$$R \equiv D \times \frac{L_{II}}{L_I}, \tag{7.182.2}$$

in which D is the distribution coefficient of the solute, and L_I and L_{II} are the flow rates of phase I and phase II, respectively. Physically, R is the ratio of the *amount* of solute in phase II to that in phase I, whereas D is the ratio of the *concentrations*. In the extraction stage, I and II are the aqueous and organic phases, respectively, and D is defined by equation (7.157.1). On the other hand, for a back-extraction or stripping operation, I and II are reversed and D is the reciprocal of that given above.

7.183. If R exceeds unity, as will shortly be seen to be necessary if extraction is to be possible, then provided n is of the order of 10 or more, R^{n+1} is large in comparison with unity, and the latter may be neglected in the denominator of equation (7.182.1). Upon taking logarithms of both sides of the resulting equation and rearranging, it is found that

$$n = \log\left[\frac{R-1}{R(x_n/x_0)}\right] - 1, \tag{7.183.1}$$

which relates the number of stages to the flow-rate ratio and the initial and final concentrations.

7.184. The quantity x_n/x_0 is the ratio of the concentration of the extracted substance in the solution leaving the column to that (in the same phase) entering it. This is sometimes referred to as the "loss" of extractable material. In order to make the loss small, that is to say, for the extraction to be efficient, it can be seen from equation (7.183.1) that R and n should be large. That the number of theoretical stages should be large is obvious, but the extraction factor R merits further consideration.

7.185. For x_n/x_0 to decrease indefinitely with increasing n, so that advantage may be taken of increasing the number of theoretical stages, it is necessary that R should be greater than unity. It is desirable, therefore, that the distribution coefficient D should be as large as possible. However, if D is less than unity the ratio of the flow rates of the liquids, i.e., L_{II}/L_I, must be adjusted so that $R > 1$, otherwise extraction of the solute from phase I into phase II will be ineffective, as shown below. It may be mentioned that, for practical reasons, the flow-rate ratio, either L_I/L_{II} or L_{II}/L_I, can rarely exceed 10 to 1, and in most cases it is in the range of 1 to 1 to 5 to 1.

7.186. If $R < 1$, the expression for x_n/x_0, as given by equation (7.182.1), approaches a limiting value equal to $1 - R$, so that the ratio x_n/x_0 approaches

unity as R becomes smaller. This means that if R is sufficiently small, x_n/x_0 will be close to unity and there will be a negligible amount of extraction, regardless of how many theoretical stages are in the extraction column. This is the condition that is desirable for the fission products in the extraction of uranium and plutonium from spent reactor fuel elements. The fission products will then tend to remain in the aqueous solution while the uranium and plutonium pass into the organic phase.

7.187. The degree of separation achieved in a given column will be greater the greater the ratio of x_n/x_0 for the desired (product) element, e.g., uranium or plutonium, to that for the undesired (impurity) elements, e.g., fission products. Good separation will result, therefore, when the extraction factor, R, for the former, which should be greater than unity, greatly exceeds R for the latter, which is preferably less than unity. This means that R(product)/R(impurity) should be as large as possible. Since the ratio of the flow rates is the same for both substances, it follows from equation (7.182.2), which defines R, that the best separation will be obtained in solvent extraction when the ratio of the distribution coefficients, i.e., the separation factor (§ 7.157), is large.

Choice of Contactor

7.188. In the design of efficient contacting equipment for solvent extraction processes involving two immiscible liquid phases, there are three important objectives; these are (1) to attain the maximum interfacial area between the two phases, (2) to permit countercurrent flow with the minimum density difference between the phases, and (3) to minimize the total volume of liquid held up by the equipment. In addition, for operation with highly radioactive solutions, it is essential that the equipment should require little or no maintenance.

7.189. Several different types of contactors have been employed for industrial continuous countercurrent liquid extractions; three in particular are in use, either on the large-scale or in pilot-plant operations, in connection with the treatment of spent reactor fuel. These are packed columns, pulse columns, and mixer-settlers; they will be described and their relative merits considered briefly.*

7.190. Taking into consideration the purpose for which it is required, the essential characteristics of contacting equipment for liquid-liquid extraction are as follows: (1) long operating life with low maintenance requirements, (2) small size in order to minimize shielding structures, (3) simplicity of operation and control, (4) easy replacement of parts, and (5) adaptability to changes in

* See, for example, V. S. Morello and N. Poffenberger, *Ind. Eng. Chem.*, **42**, 1021 (1950); B. V. Coplan, J. K. Davidson, and E. L. Zebroski, U. S. Patent 2,646,346 (1953), Report AECU-2639; *Chem. Eng. Prog.*, **50**, 403 (1954); M. W. Davis, Jr., T. E. Hicks, and T. Vermeulen, Report AECD-3545; J. D. Thornton, *Chem. Eng. Prog. Symposium Series*, No. 13, **50**, 39 (1954); G. Sege and F. W. Woodfield, *ibid.*, No. 13, **50**, 179 (1954); *Chem. Eng. Prog.*, **50**, 396 (1954).

process conditions. The three forms of contactor considered below are generally satisfactory in these respects.

7.191. Packed columns are filled with some form of packing material, such as Raschig rings. These serve to break up the organic liquid into droplets of dispersed phase which are compelled to follow a tortuous path through the continuous (aqueous) phase. As a result, contact between the two phases is enhanced. At the top of the column there is a clear space where the droplets coalesce and are drawn off as a continuous phase.

7.192. A pulse column has a number of finely perforated plates placed across the tower. By means of a reciprocating piston, acting as a pulse generator, at the bottom of the column, the liquids are forced through the holes in the plates in fine droplets. The turbulence produced by the pulses results in very effective contact between the two liquid phases. The efficiency is greater than that of a packed column and the same degree of extraction can be achieved in a shorter tower.

7.193. A mixer-settler stage consists of two sections; as the name implies, one is a mixer and the other is a settler, or separator, as illustrated diagrammatically in Fig. 7.17. In the mixing part the two phases are brought into intimate contact by passage through fine orifices or by means of an agitator. The liquids then pass into the settling part of the stage where the phases separate into two continuous layers. The liquid streams then flow in opposite directions to two

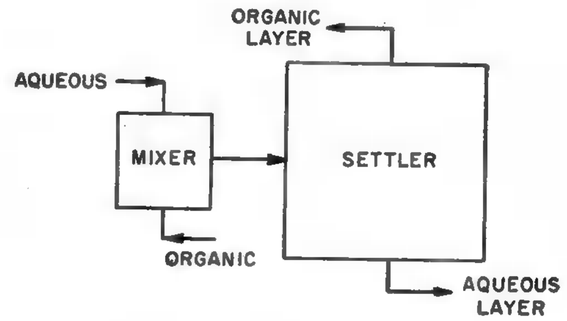


Fig. 7.17. Block diagram of one stage of mixer-settler unit

other mixer-settler stages, thus providing a countercurrent extraction system. Each mixer-settler stage has an efficiency of about 90 per cent of theoretical. Consequently the number of stages required to effect a prescribed degree of extraction is 10 per cent or so larger than the calculated number of theoretical stages, using equation (7.183.1).

7.194. The chief advantage of the mixer-settler system over a column is that in most forms little headroom is required, although a fair amount of floor space may be necessary. The height of the equipment is especially important in connection with the processing of radioactive solutions since the structures

needed for shielding of tall columns are expensive. The disadvantage of columns in this respect may be overcome by the so-called "concatenated" pulse column. The required column length is divided into several sections which are mounted in a rack and connected in series by a suitable piping arrangement. The equivalent of a 50-ft tall column may thus be installed in a one-story building with about 10 to 12 ft of headroom.

7.195. At the present time no contactor appears to be overwhelmingly superior to all others, and the choice, usually between pulse columns and mixer-settlers, is determined largely by circumstances and individual preference. Among the other types of contactors being considered, mention may be made of the centrifugal contactor, in which centrifugal force is used to bring the liquids into intimate contact. It appears to have certain advantages, e.g., low hold-up, high flow capacity, and small space requirements, as compared with other contactors.

OTHER SEPARATIONS PROCESSES

Liquid-liquid Extraction by Fused Salt

7.196. In addition to the separations procedures already described, there are one or two other methods, still in what might be referred to as the development stage, that deserve mention. One of these is a liquid-liquid extraction process of an unusual character that has been considered for the treatment of the (dilute) solution of uranium in molten bismuth which is the fuel for the proposed Brookhaven Liquid Metal Fuel Reactor (see § 12.141). The extractant is a fused eutectic mixture of lithium and potassium chlorides at 450°C (840°F). When brought into equilibrium with the liquid uranium-bismuth alloy containing rare-earth elements, such as might be produced in fission, it is found that the rare earths are extracted almost completely into the fused-salt phase whereas the uranium remains in the bismuth phase. It is probable that other fission-product elements would be removed at the same time, so that extraction with fused chlorides would appear to present an interesting possibility for the processing of spent reactor fuel in certain circumstances.*

7.197. It should be pointed out that the extraction with a fused salt is more closely related to the familiar "slagging" of metals than to the extraction by an organic solvent described earlier. The rare-earth element lanthanum, for example, does not dissolve as such in the fused chloride, but it first reacts chemically with the lithium chloride, thus

$$\underset{\text{(metal phase)}}{La} + \underset{\text{(salt phase)}}{3LiCl} \rightarrow \underset{\text{(salt phase)}}{LaCl_3} + \underset{\text{(metal phase)}}{3Li}.$$

* D. W. Bareis, R. H. Wiswall, Jr., and W. E. Winsche, *Chem. Eng. Prog. Symposium Series*, No. 12, 50, 228 (1954); C. Raseman and J. Weisman, *ibid.*, No. 12, 50, 153 (1954). Somewhat shorter versions of these reports appear in *Nucleonics*, 12, No. 7, 16, 20 (1954).

The resulting lanthanum chloride ($LaCl_3$) then dissolves in the fused salt whereas the elemental lithium produced in the reaction passes into the metal (uranium-bismuth) phase. (There is also a similar reaction with the potassium chloride but to a much smaller extent.) The extraction thus involves essentially the removal of the lanthanum (or other metal) dissolved in the bismuth and its replacement by lithium.

7.198. Theoretical considerations indicate that the efficiency of the extraction of any element from the bismuth phase should increase with the stability of the chloride of the element. It is because of the relatively low stability, i.e., small (negative) free energy of formation per chlorine atom, of uranium tetrachloride, as compared with that of the rare-earth chlorides, that uranium is not extracted by the fused chloride whereas the rare-earth elements are extracted. For the same reason, namely, the comparative instability of bismuth trichloride, the liquid bismuth acts essentially as an inert solvent for the metals and does not react appreciably with the lithium chloride.

7.199. Although there is a possibility of using fused chlorides for separating rare-earth (and perhaps other) elements from uranium, the procedure described above is not satisfactory for its intended purpose, the treatment of the uranium-bismuth alloy fuel. The reason is that the large neutron absorption cross section of the lithium makes the liquid bismuth after extraction unsuitable as a fuel medium for a thermal reactor. In addition the lithium chloride-potassium chloride mixture is very sensitive to the presence of traces of oxidizing or reducing impurities which can completely upset the extraction. These difficulties may be overcome by the use of another chloride in place of lithium chloride and by control of the oxidation-reduction state of the system.

Separation by Volatilization

7.200. The separation of elements, by making use of differences in vapor pressure of the elements themselves or of suitable compounds, has been employed for many years, especially in the study of radioactive materials. The possibility of extracting plutonium from spent reactor fuel by volatilization was considered in the early stages of the atomic energy program, but this separations method was not pursued in detail because the precipitation and solvent extraction procedures seemed to offer greater prospects of immediate success. However, for certain reactor applications, as will be indicated below, the volatilization method may prove attractive.

7.201. An examination of the familiar properties of the simpler compounds of uranium shows that the hexafluoride (UF_6) is a readily volatile solid (see § 8.27), whereas the trifluoride (UF_3) and the tetrafluoride (UF_4) are nonvolatile. Presumably the same differences in volatility occur in the corresponding oxidation states of the other actinide elements, so that a method of separation appears feasible, at least in principle. The fact that the fluorides of many fission prod-

ucts, e.g., the rare-earth elements, are essentially nonvolatile is, of course, advantageous from the standpoint of the treatment of spent reactor fuel.

7.202. A possible application of the volatilization method is in connection with the continuous processing of a thorium breeder blanket. The thorium-232 captures neutrons and the product then undergoes two stages of beta decay to form, first, protactinium-233 and then uranium-233 (§ 1.61). It would be advantageous if these two latter isotopes could be removed continuously because losses due to neutron capture could thus be minimized. In the method proposed for achieving this end the thorium blanket would be in the form of the nonvolatile solid thorium tetrafluoride (ThF_4), and a fluorine-containing gas would be passed through it at high temperature. The fluorine would then react with the protactinium and uranium to form the volatile fluorides, PaF_5 and UF_6, respectively, but the thorium tetrafluoride would be unaffected. Consequently the protactinium-233 and uranium-233 would be removed in the stream of gas but the thorium would remain. Experimental investigations have shown that the method is possible, but difficulties arise from the sintering of the thorium fluoride powder and the accompanying decrease in the rate of diffusion of gas through the blanket. It is not certain that these difficulties can be overcome because of the narrow temperature range within which effective operation appears feasible.*

PROCESS WASTE DISPOSAL†

Liquid Wastes

7.203. The chief waste disposal problem associated with reactor fuel processing is concerned with the handling of the fission product residues. This problem has hitherto been solved in such a manner as not to constitute a hazard either in the chemical plant itself or in the surrounding areas. It must be admitted, however, that the solution is not altogether satisfactory from the economic standpoint, and studies are in progress with the object of finding improved methods of waste treatment. For convenience, the problems of disposal will be discussed briefly here according to the physical form, i.e., gas, liquid, or solid, of the waste material.

7.204. Since the liquid wastes constitute the major aspect of the disposal problem, these will be considered first. Because of the potential value of mixed or of individual fission products (§ 7.208) and also because their intense radioactivity constitutes a potential health hazard, the reactor fuel wastes cannot be treated in the same manner as are the waste products of other industries. Consequently most waste solutions from fuel processing plants, possibly after some

* F. T. Miles *et al.*, *Chem. Eng. Prog. Symposium Series*, No. 12, **50**, 173 (1954); *Nucleonics*, **12**, No. 7, 26 (1954).

† The discussion of radioactive waste disposal is here restricted to certain aspects associated with fuel processing. A more complete treatment of the over-all problem is given in Chapter IX.

treatment, are stored in underground tanks. This is not a very desirable procedure for several reasons. The volume of storage space is continually increasing, and there is a constant risk of leakage. In addition, the absorption of the beta and gamma rays may result in undesirably high temperatures being attained in solutions having considerable activity.

7.205. Solutions containing fertile and fissionable materials, i.e., uranium, thorium, and plutonium, are usually stored separately from those containing only fission products. Where possible, the solutions are first evaporated to decrease their volume, so as to minimize the space required for storage. In some cases a useful process chemical, e.g., nitric acid, may be recovered in the course of evaporation.

7.206. The large volumes of waste solutions having a low level of radioactivity, e.g., aqueous solutions obtained in later stages of an extraction process, are also treated separately. Such solutions have been disposed of by discharge into dry wells or in open pits in the ground. The liquids slowly seep out into the surrounding soil where the radioactive ions are adsorbed. By this means, permanent disposal of large volumes of wastes of low activity can be achieved. There is little danger of the spread of radioactivity unless the adsorption capacity of the soil is exceeded and the waste solution enters a water table in general use. This possibility may be avoided by careful evaluation of the site geology before utilization of this method of waste disposal.

7.207. Another procedure that has been used in connection with treatment of the large volumes of dilute solutions is to concentrate them by evaporation. The small residual volumes can then be stored while the condensate, if of sufficiently low activity, can be allowed to mix off with large volumes of water in a river. Certain fission products, notably iodine and ruthenium, may volatilize during distillation and hence appear in the condensate, thus increasing its radioactivity. The iodine, at least, can be retained by making the solution weakly alkaline before evaporation.

7.208. If it were not for the possible ultimate value of the fission products, more effective methods of concentration and final disposal of wastes would perhaps be used. Several procedures have been worked out (see § 9.129), which may have to be employed, in due course, if the storage problem becomes acute. In any case, if the solutions are kept for several years, some of the desirable fission products will be lost as a result of radioactive decay and their possible economic value will decrease. In this event, further storage may prove unnecessary.

Gaseous Wastes

7.209. There are three important radioactive waste gases, namely, iodine and the chemically inert gases krypton and xenon. These are liberated when the fuel element is dissolved. If nitric acid is used for this purpose the gases are diluted with large volumes of oxides of nitrogen. The gases from the dissolver

vessels are generally discharged to the atmosphere through high stacks (Fig. 7.18). Provided the spent fuel has been cooled for about 100 days or more, to permit appreciable decay of the iodine-131 (see Fig. 7.4), the gases disposed of in this manner constitute no significant hazard. Continuous observations are made of the meteorological conditions to make sure that there is no tendency for the gases expelled from the stacks to return to the earth's surface.

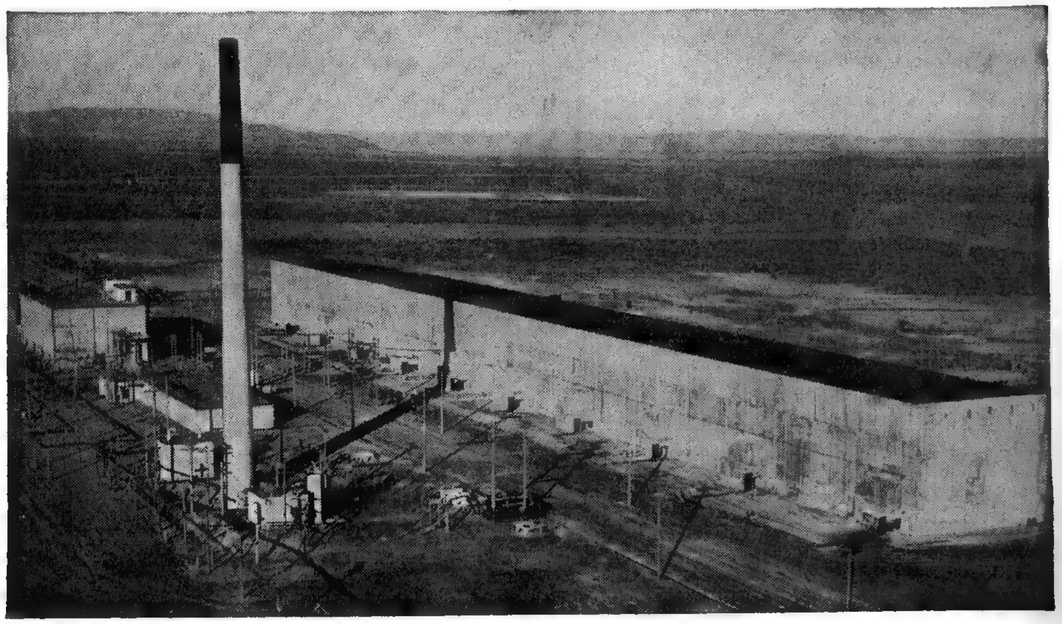


FIG. 7.18. Spent fuel processing plant at Hanford, Washington

7.210. An alternative method of disposal of the radioactive gases is possible when for meteorological, or other, reasons discharge into the atmosphere is undesirable. The iodine and oxides of nitrogen could be removed by scrubbing the gases with a dilute solution of alkali, and the krypton and xenon could be adsorbed on carbon or silica gel. The resulting solution and solid, respectively, would then have to be disposed of in a suitable manner.

7.211. Under the heading of hazardous gaseous wastes may be included air from processing plants carrying small solid or liquid radioactive particles. Since this may constitute a serious danger, the air is discharged through filters of sand or, better, of glass-wool to remove the particulate matter. These filters have a limited operating life and must be replaced periodically. The spent filter represents a problem in the disposal of solid waste, to which reference will be made below.

7.212. The control and segregation of air, from operations where its contamination is to be expected, are desirable in order to minimize the volume of air that must be treated. Thus in chemical processing plants the cells and process vessels are usually maintained under reduced pressure to eliminate the possibility of radioactive particles escaping into the air. By keeping the leakage as low as possible, the volume of contaminated air is greatly decreased. As a general rule, therefore, it is necessary to filter only the limited volumes of air from the process vessels.

SOLID WASTES

7.213. The solid radioactive waste from a fuel processing plant may consist of such items as discarded process equipment, which has become contaminated by contact with active solutions, and filter material, mentioned above. If the equipment is worth recovering, it may frequently be decontaminated to an extent that will make repair possible. For stainless steel, which is extensively used in fuel processing, various acid solutions can be used as decontaminating agents. Mild steel vessels are difficult to decontaminate because they are attacked and corroded by acids, but there is a possibility that they may be used as scrap in blast-furnace operation, when the radioactivity will be removed in the slag.

7.214. When decontamination of a solid is not possible or desirable, the material is usually buried in the ground. Consideration is being given to marine burial as a method of permanent disposal of solid radioactive wastes. This procedure is being used, to a limited extent, on both the Atlantic and Pacific coasts, but for plants in the interior of the United States the cost of shipping the materials may be prohibitive.

USES OF FISSION PRODUCTS

7.215. The utilization of fission products is in a sense an aspect of the waste disposal problem. If extensive industrial uses could be found, either for certain individual fission products or for the complex mixture obtained in spent reactor fuel treatment, the disposal situation would be greatly changed. If there is to be a real future for nuclear energy, it would appear that uses will have to be found for the fission products. Several investigations, sponsored by the U. S. Atomic Energy Commission, have been or are still being made for the purpose of finding possible applications for these substances. Some of these applications are summarized in Table 7.6;* they are mostly dependent upon the ionization produced by the beta and gamma radiations.

* "Industrial Uses of Fission Products," Project 361, Stanford Research Institute; Report AECU-1673; *Nucleonics*, **10**, No. 1, 45 (1952); University of Michigan Project, Reports COO-124, COO-196, COO-198, AECU-2981; Vitro Corp. of America, Report KLX-381; D. Duffey, *Nucleonics*, **11**, No. 10, 8 (1953); L. E. Crean, *et al.*, *ibid.*, **11**, No. 12, 32 (1953); B. Manowitz, *ibid.*, **11**, No. 10, 18 (1953), *Chem. Eng. Prog. Symposium Series*, No. 12, **50**, 201 (1954).

TABLE 7.6. POSSIBLE INDUSTRIAL USES OF FISSION PRODUCTS

Application	*Remarks*
Activation of phosphors	For luminescent pigments for use in signs visible in the dark. Very pure isotopes necessary.
Static eliminators	Ionization of air allows static charge to leak away. May also prevent agglomeration of fine particles in grinding processes. Useful in printing, textile, plastic, and rubber industries.
Fluorescent light tubes	Permanent ionization of the gas will permit faster starting and lower starting voltages.
Instruments	Thickness and level-indicating gauges based on penetration of beta or gamma rays. Marking of interface between products in pipelines.
Radiography	Gamma-ray sources of high activity and low energy give sharp definition.
Cold sterilization of foods and drugs	Beta and gamma rays destroy microorganisms. Especially advantageous for heat-sensitive products, e.g., antibiotics. Prevent off-taste flavors in canned foods; increase shelf life of fresh fruits and vegetables.
Power source	Direct conversion of beta-particle energy into electricity, e.g., in a battery. Pure beta-emitter of relatively long half life desirable, e.g., strontium-90.
Heat source	Because of self-absorption of beta and gamma rays, radioactive material has higher temperature than surroundings.
Chemical reactions	Facilitation of flame propagation in internal-combustion and jet engines; polymerization, etc.

DESIGN OF RADIOCHEMICAL PROCESSING PLANTS *

Stages in Reactor Fuel Processing

7.216. The design of a radiochemical processing plant for the treatment of spent reactor fuel may be based on the following stages:

1. Storage (or cooling) of the irradiated material to permit decay of short-lived fission products and intermediate species, e.g., neptunium-239.
2. Dissolution of the fuel elements and preparation of the feed solution.
3. Storage of feed solution prior to introduction into the separation process.
4. Separation of fissionable and fertile materials from fission products.
5. Collection and disposal of the waste streams containing fission products.
6. Collection of product streams and preparation for shipment.
7. Preparation, distribution, and recycling of process reagents.

7.217. From a general engineering standpoint, there is no essential difference between the operation of a plant for processing reactor fuel and that of other chemical plants. However, there are certain special factors involved in reactor fuel treatment which complicate both design and operation and increase capital and operating costs. They are chiefly (1) the highly radioactive nature of the materials to be processed and (2) the possibility that solutions of fissionable species may become supercritical and so permit the propagation of a divergent fission chain reaction.

* For description of pilot plant, see H. K. Jackson and J. W. Ullmann, Report AECD-3338; also, C. M. Nichols and A. S. White, *Chem. Eng. Prog. Symposium Series*, No. 13, **50**, 129 (1954).

7.218. In order to provide protection for operating and maintenance personnel from radiation, adequate shielding must be provided. Except for a short time after removal of the fuel element from the reactor, when delayed neutrons are still being produced, there is no neutron shielding problem. Further, although uranium and plutonium emit alpha particles, their range is so small that they may be disregarded. Hence, as far as the process equipment is concerned, shielding is necessary for beta and gamma rays, including the bremsstrahlung accompanying the deceleration of beta particles. Actually the problem is reduced to protection from gamma rays (and bremsstrahlung) from fission products since these are more penetrating than beta particles. Shielding which attenuates gamma (and X-) radiation to harmless proportions will automatically be effective against alpha and beta particles.

7.219. The most common gamma-ray shielding materials, in order of increasing density, are water, concrete, iron, and lead. As seen in § 2.68, the thickness of material required to produce a certain degree of gamma-ray attenuation is approximately inversely proportional to its density. A lead shield will consequently occupy a smaller volume than one made of any of the other three common shielding materials. Because of its high cost, however, lead is used only where space is at a premium or where more bulky shields are not practical.

7.220. Water provides the least expensive shield, although a thickness of several feet may be required. It is used where its liquid nature is not a drawback. For example, after removal from a reactor, the irradiated fuel elements are stored under 12 to 20 ft of water in a large basin, often referred to as a "canal." The water serves not only as a radiation shield but also as a means for thermally cooling the fuel elements. Radioactive decay and the absorption of the emitted radiations are accompanied by considerable heat evolution.

7.221. All process vessels, equipment, and lines containing or carrying radioactive liquids or gases, e.g., contaminated air, must be shielded. In special cases, lead or iron may be used as shield material, as mentioned above, but, for bulk shielding, concrete is used. The thickness of such shields may range from 1 to 6 ft. With necessary pipe inserts, the shields are expensive, but it is sometimes possible to make them serve a dual purpose by integration into the structure of the building. Bulk shielding costs can be reduced by limiting the number of processing units, e.g., by continuous rather than batch operation, and by making the space between vessels as small as is compatible with other requirements, e.g., maintenance, criticality, etc.

Maintenance of Equipment

7.222. Equipment used for radioactive solutions must be properly designed and manufactured so as to require the absolute minimum of repair or replacement. Special precautions must be taken to ensure good workmanship, and the materials of construction should be resistant to the process solutions. All piping must be absolutely leaktight. The use of complicated equipment with moving parts requiring lubrication should be avoided if possible. The same stipulation applies to valves and pumps with packing, because the packing, when replaced due to normal deterioration, would be contaminated. If equipment that might need fairly frequent maintenance must be used, then it should be installed in separate shielded compartments.

7.223. In spite of all precautions taken to prevent equipment failure, it is inevitable that some maintenance will be required. In the design of radiochemical processing plants, two basically different approaches have been used; they are (1) remote maintenance and (2) direct maintenance, which will be reviewed in turn.

*Remote maintenance**

7.224. The equipment and piping are so designed that when repair is necessary any item can be removed and replaced by a duplicate using remotely operated mechanical devices. The faulty equipment can then either be discarded or decontaminated and repaired elsewhere. The remote-maintenance plant consists of a long, high building, called a "canyon" (see Fig. 7.18). A considerable amount of space is required since every piece of equipment must be located in such a manner that it can be reached by a crane without displacing any other piece.

Direct maintenance

7.225. The equipment and processing cells are designed so that they can be decontaminated, by flowing suitable solutions through them, to an extent that will permit maintenance personnel to effect direct repair in place. In the direct-maintenance plant, cells with concrete walls are used to segregate process equipment according to function and activity. Isolation of blocks of equipment makes possible the decontamination and repair of a section of the plant while other sections remain in operation. The installation is determined largely by process and engineering requirements, so that the plant is more compact than the remote-maintenance canyon.

7.226. In reviewing the various aspects of the two types of processing plants, it would appear that the chief advantage of the remotely maintained plant is that it permits operation with the minimum of interruption. Even a major break can be rectified in a short time by removal and replacement of the equipment. Direct maintenance, on the other hand, may mean that in an emergency

* W. M. Harty, *Chem. Eng. Prog. Symposium Series*, No. 18, 50, 115 (1954).

the plant will be shut down for a period of time sufficient to accomplish both decontamination and repair. However, where duplicate plant units have been installed, maintenance can be carried out without shut-down. The canyon type of construction also makes possible the use of especially large pieces of equipment, e.g., vessels holding very large volumes of solution, which are not easily accommodated in a cell.

7.227. The disadvantages of remote maintenance are largely economic. A much larger canyon is required so that both capital and operating costs are greater than for direct maintenance. In addition, much greater effort is demanded in the design of the plant so that every piece of equipment and piping can be readily removed by remote control. Although in direct maintenance there is some duplication of important equipment, remote maintenance requires a large stock of spares for all units. Finally, it is considered that the remote maintenance type of design means less flexibility and that process changes are more difficult to incorporate than in directly maintained plants.

7.228. At this time it is not possible to state definitely that the advantages of either type of design so greatly outweigh its disadvantages as to make it preferable to the other. Remote-maintenance plants seem to be more desirable where large volumes of solution must be treated and where continuity of production is important. On the other hand, the lower construction and operating costs of a direct-maintenance plant may be the determining factors when smaller volumes are to be processed.

Avoidance of Criticality

7.229. An aqueous solution of a fissionable material may become supercritical under certain conditions of concentration and size and shape of the containing vessel. Such conditions might easily arise in a processing plant unless special efforts are made to avoid them. Although a supercritical solution would probably not explode, the rapid evolution of large amounts of heat would undoubtedly result in an unpleasant accident. Consequently precautions against the attainment of criticality must be adopted in the design and operation of the chemical plant.

7.230. Three types of action are taken to avoid criticality. First, the amount of relatively pure fissionable material that may be present in any one vessel is definitely limited. Since the concentrations of the solutions are known at all stages of the separation process, the vessels are designed so that their contents will not become critical. There is a chance, however, that the normal concentrations may be exceeded due to an operational error, and so a considerable safety factor is included in the calculations.

7.231. A second aspect of plant design is the possibility of using so-called "always safe" equipment. Consider, for example, a cylindrical vessel containing a solution of fissionable material. As seen in Table 3.7, the critical height, H, and radius, R, are related by

$$B^2 = \left(\frac{2.405}{R}\right)^2 + \left(\frac{\pi}{H}\right)^2,$$

where B^2 is the buckling of the critical system. For a solution of specified composition, B^2 has a definite value, and so, as the radius of the cylinder is decreased, the critical height must increase. However, when R is equal to $2.405/B$, it is seen that H would have to be infinite. In other words, for every solution of fissionable material, there is a certain radius below which a cylindrical vessel will never become critical no matter what its height. Such a vessel would be "always safe" for the given solution. A change in concentration would, however, result in a change in the buckling (see Fig. 3.13) and the vessel could become critical.

7.232. The third consideration in the avoidance of criticality is the proper spacing and location of units. Thus, if two "always safe" cylinders were placed sufficiently close together, the combined system might quite possibly become critical.* Vessels containing moderately concentrated solutions of fissionable material must consequently be separated by an adequate distance. At present, safe criticality practice is determined largely empirically, and design standards can be fixed more definitely only as further experimental facts are accumulated.

Process Plant Instrumentation†

7.233. Since most of the plant units are inaccessible, remote instrumentation is necessary, both for control and sampling purposes. Apart from instrumentation for the standard service facilities, instruments are used mainly for the following process measurements: temperature, liquid level, specific gravity, fluid flow rate, and pressure. The control room of a radiochemical processing plant is depicted in Fig. 7.19.‡

Temperature

7.234. The device most commonly employed for temperature measurement is the thermocouple. Its small size permits its installation in many places where more bulky detectors could not be used. Thermocouples have a fast response to temperature changes and are available in wide variety. In conjunction with an indicating and recording instrument at a distance, temperatures can be read with adequate accuracy. By employing an electronic-type recorder, thermocouple leads may be run for considerable distances without introducing appreciable errors. In many reactor vessels a thermocouple combined with a recording-controlling instrument provides automatic temperature control.

Liquid Level and Density

7.235. These two quantities are generally determined by means of a pneumatic purge or "bubbler" system. An open-ended tube, called a dip tube, is

* Cf. R. L. Macklin, Report AECD-3274.
† Cf. V. L. Parsegian, *Nucleonics*, 6, No. 1, 38 (1950).
‡ Oak Ridge National Laboratory (private communication).

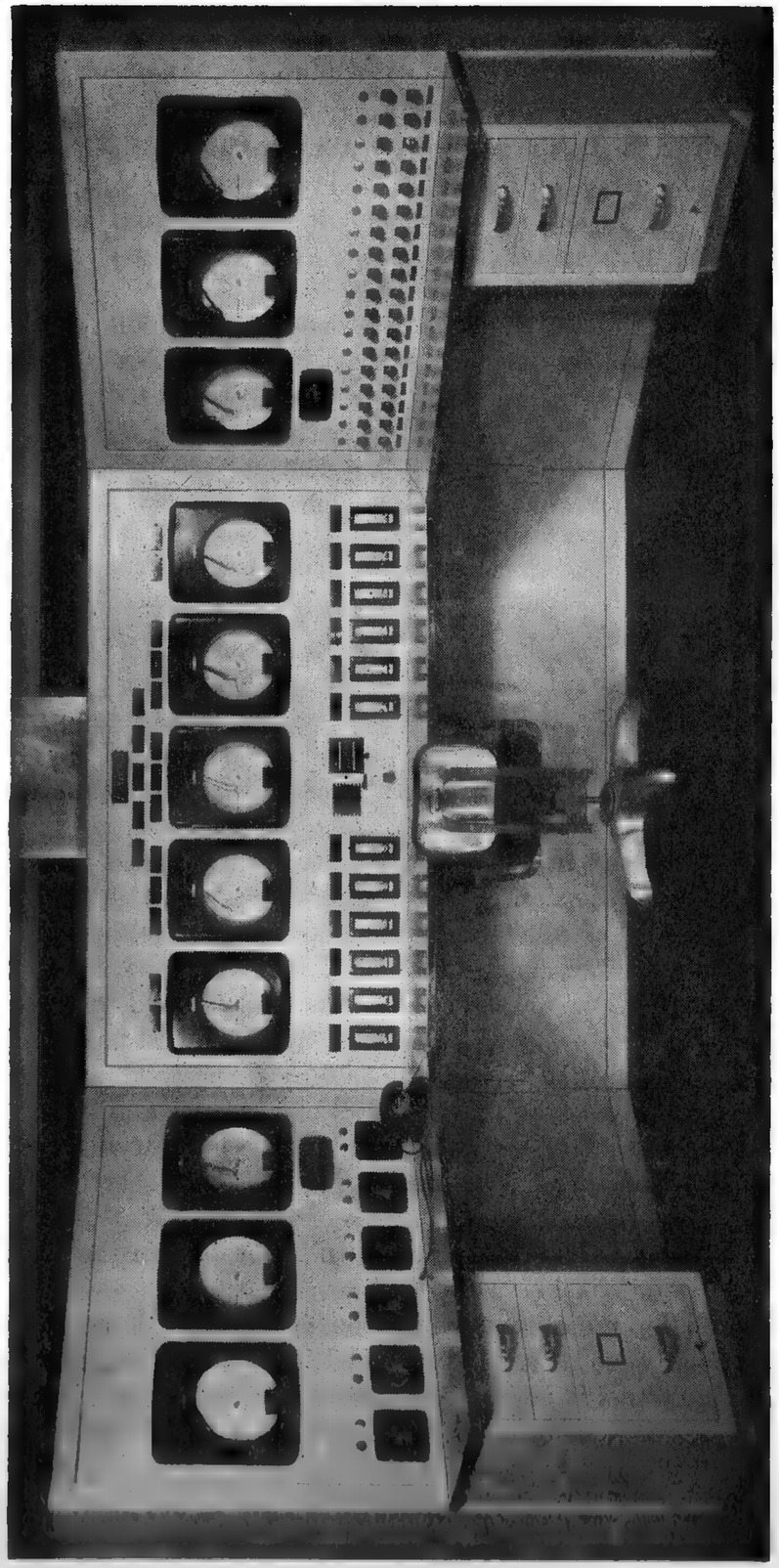


Fig. 7.19. Control room of radiochemical processing plant (Oak Ridge National Laboratory)

fixed vertically in the vessel containing the liquid. A supply of clean compressed gas, e.g., air, nitrogen, etc., is attached to the top of the dip tube, and the pressure required to discharge the gas from the lower end of the tube is recorded by a manometer. This pressure is clearly a measure of the hydraulic head in the vessel and hence is proportional to the liquid level. By using a suitable pneumatic transmission line, the pressure (and level) indicator and recorder can be placed at a distance from the vessel.

7.236. In order to determine the density of the liquid, two dip tubes are used, their open (lower) ends being separated by a known vertical distance. The difference in the pressures in the two dip tubes is equal to the hydraulic head between the two orifices. Since the orifices are a fixed vertical distance apart, the differential pressure is a measure of the density of the liquid. The recording instrument can be calibrated to read the density directly.

Fluid Flow Rate

7.237. A simple and reliable method for determining flow rates is to observe the rate of fall of liquid level in the feed tank. When this is not possible or when continuous recording is required, a flow meter is employed. In the variable-head type of meter, the difference in pressure is determined on the two sides of an orifice or Venturi restriction placed in (or parallel with) the flow stream. The advantage of the Venturi tube lies in the fact that the shape of the restriction permits its use in solid-bearing streams. A pneumatic transmitter is usually employed to indicate the differential pressure, and hence the flow rate, on a distant recorder. It should be mentioned that flow meters of the orifice or Venturi type are not too reliable and require frequent calibration.

7.238. In the variable-area flow meter (or Rotameter) the differential pressure is held constant and the area of the restriction is allowed to vary. This is achieved by means of a cone-shaped vertical tube and a metal float or bob which is free to move inside the tube in response to flow changes. For every vertical position of the float, there corresponds a definite orifice area and flow rate for the fixed pressure difference. The only force pulling the float downward is that of gravity and this is constant. The area meter has a number of advantages, especially for low flow rates. Since the area of the opening is directly proportional to the flow rate, the scale is linear, which is not true for the variable-head meter.

7.239. A method for remote determination of the flow of good electrically conducting solutions, such as are encountered in process chemistry, by means of electromagnetic meters is being investigated. If the liquid flows through a pipe placed in a magnetic field, an electromotive force is induced in the moving stream and can be picked up by two electrodes inserted into opposite sides of the pipe. Since the electromotive force is small, it is amplified electronically and recorded. Because of its high cost, the electromagnetic flow meter is not

in general use. Its main application, at present, is in connection with more difficult radioactive streams or low flow rates.

Pressure

7.240. Pressure determinations in separation plants are usually limited to the measurement of a few feet of water above or below atmospheric pressure. Various types of manometers can be used with either pneumatic or electrical transmission systems to permit remote recording.

Conclusions

7.241. In the preceding paragraphs an attempt has been made to outline some of the procedures adopted to deal with the unusual situations arising from the necessity for handling, on the industrial scale, materials which are not only highly radioactive but have, in addition, the potentiality of sustaining a fission chain reaction. Because of the special design and construction required to safeguard personnel and to permit continuity of operation, the processing plants are so complex as to defy adequate description in the limited space available here.

Symbols Used in Chapter VII

B^2	buckling
C	number of free volumes of eluent for maximum ion concentration
D	homogeneous distribution coefficient (precipitate/solution)
D	distribution coefficient (organic phase/aqueous phase)
d	designation of electron subgroup
E	oxidation-reduction potential
E_f	formal potential
e^-	electron
e	base of natural logarithms
F	number of free volumes
f	designation of electron subgroup
f_c	fraction of carrier element in precipitate
f_t	fraction of trace element in precipitate
H	height of critical cylinder
K_d	distribution coefficient for ion exchange
k_c	distribution of carrier element between precipitate and solution
k_t	distribution of trace element between precipitate and solution
L	flow rate of liquid
n	difference in charge of oxidized and reduced states
n	number of charges on ion
n	number of theoretical stages (or plates)
p	designation of electron subgroup
R	radius of critical cylinder

R extraction factor
s designation of electron subgroup
T absolute (Kelvin) temperature
T_0 time of reactor operation
W half width of elution curve (in free volumes)
x_0 initial concentration of solute
x_n final concentration of solute (after n stages)
α separation factor

PROBLEMS

1. A reactor containing 7 lb of highly enriched uranium as fuel operates at a power of 30,000 kw continuously for 100 days. The fuel is then removed and allowed to cool for 80 days. Estimate (*a*) the activity in curies per pound of fuel and (*b*) the activity in curies per pound of fission products.

2. In the preceding problem, suppose that at the end of the 80-day cooling period the barium is rapidly separated from the fission products. What would be the approximate activity in curies per pound, assuming it to be due entirely to barium-140?

3. Account for the fact that in Fig. 7.4 the fractional activity for some isotopes increases with time after removal from the reactor whereas for others it decreases steadily. Would this difference in behavior be expected to continue indefinitely?

4. Outline a scheme for the extraction of plutonium and the recovery of uranium from spent fuel elements by the ion-exchange method.

5. In an ion-exchange process, using a resin column 1 meter long, the "half width" of the elution curve for a given ion was found to be 0.94 free volume and the concentration of the ion in the eluent was a maximum when 6.7 free volumes had passed through the column. Estimate the average height equivalent to a theoretical stage.

6. It is required to extract a particular solute from an aqueous solution, containing a salting agent, using an organic solvent; the maximum permissible "loss" is to be 1 part in a million. The flow rate of the organic liquid is 2.4 times that of the solution, and the distribution coefficient of the solute between organic and aqueous phases under the given conditions is 0.55. What is the minimum number of actual mixer-settler stages that would be needed?

*Chapter VIII**

NUCLEAR REACTOR MATERIALS

REACTOR FUELS

Introduction

8.1. Many of the materials used in the construction of a nuclear reactor are the same as those employed in other forms of engineering construction. However, the characteristics of the fission chain reaction necessitate the use of a number of special materials which have not hitherto been familiar in industry. The materials required for reactor construction may be divided into classes according to their particular function; they are (1) fuel, (2) moderator and reflector, (3) structure and cladding, (4) coolant, and (5) shield.† The last of these categories will be treated in a later chapter in connection with the shielding aspects of reactor design; the others will be discussed here.

8.2. In a reactor fuel there must be present at least one of the fissionable species, i.e., uranium-233, uranium-235, or plutonium-239. These are derived from two naturally occurring source materials, namely, uranium and thorium. The former normally contains 0.7 per cent of the fissionable uranium-235, and, in addition, the nonfissionable uranium-238 can be converted into plutonium-239. Thorium has no fissionable isotope, but uranium-233 can be obtained from it by appropriate nuclear reactions. Consequently the consideration of reactor fuels may well begin with an account of the sources, production, and properties of the elements uranium and thorium.

Sources of Uranium‡

8.3. Uranium is by no means a rare substance; it has been estimated that it is present to the extent of about 4 ppm of the earth's surface. This means that

* Material contributed by K. B. Brown, G. H. Cartledge, G. E. Evans, S. Glasstone, J. R. Johnson, N. F. Lansing, S. Peterson, W. H Sullivan, and J. M. Warde.
Reviewed by K. B. Brown, E. J. Boyle, D. S. Billington, R. B. Briggs, G. A. Garrett, J. L. Gregg, W. R. Grimes, C. J. Hochanadel, J. R. Johnson, F. Kerze, Jr., R. N. Lyon, H. F. McDuffie, E. C. Miller, C. H. Secoy, and J. M. Warde.

† For general reviews, see *Nucleonics*, 11, No. 6, 18–34 (1953); S. McLain, *Chem. Eng. Prog.*, 50, 240 (1954).

‡ J. J. Katz and E. Rabinowitch, "The Chemistry of Uranium," Part I, National Nuclear Energy Series, Div. VII, Vol. 5, McGraw-Hill Book Co. Inc., 1951, Chapter 3; R. D. Macdonald, *Chem. Eng. Prog. Symposium Series*, No. 11, 50, 60 (1954).

uranium is more abundant than relatively familiar elements such as silver, mercury, bismuth, and cadmium. The total weight of uranium in the earth's crust, to a depth of 12 miles, is probably of the order of 10^{14} tons. But this figure is apt to be misleading because most deposits are of such low grade, containing 0.001 per cent or less of uranium, that extraction of the metal would appear to be uneconomic.

8.4. Of the uranium sources available to the United States, only two consist of relatively high-grade ores; these are the deposits in the Belgian Congo and in Canada. The ore as mined contains from 1 to 4 per cent of uranium, chiefly in the form of one of the primary minerals *pitchblende* or *uraninite*. These are both oxides of variable composition ranging between UO_2 and U_3O_8, so that they are represented by the general formula xUO_2, yUO_3, the ratio y/x varying from zero to two.

8.5. Uranium ores of medium grade occur in many parts of the world, including Canada and Australia. The principal deposits within the United States are those of the Colorado Plateau, an area which includes portions of Arizona, Colorado, New Mexico, and Utah. Although there are considerable variations in grade and composition, the majority of the ores mined in this region contain from 0.1 to 0.5 per cent of uranium.* Many minerals are present in the Colorado Plateau, but most of the production has been of secondary minerals, typically of the *carnotite* ($K_2O \cdot 2UO_3 \cdot V_2O_5 \cdot xH_2O$) and *autunite* ($CaO \cdot 2UO_3 \cdot P_2O_5 \cdot xH_2O$) families. Some deposits of primary uranium minerals also have been located and developed.

8.6. The carnotite ores of the Colorado Plateau often contain other vanadium minerals. These were formerly mined for vanadium, and the residues, sometimes referred to as "domestic ore concentrate," have been used as a source of uranium. At the present time both the uranium and the vanadium are extracted, as described below (§ 8.12). If the demand is sufficient the value of the vanadium may compensate to some extent for the cost of recovering the uranium. Similarly, uraniferous copper ores, occurring in the White Canyon district near Hite, Utah, have been processed for both uranium and copper. The gangues in which uranium minerals are found vary widely; they include sandstones, limestones, clays, and asphaltites. The nature of the gangue, as well as the character and grade of the uranium mineral, determine the type of processing that is applicable.

8.7. As the ores of higher uranium content are depleted, low-grade sources of the element will become of increasing significance. Among these the most immediately important are the South African (Rand) gold-ore residues, containing about 0.02 per cent of uranium, of which large amounts of fairly constant composition are available. The mining costs have already been assigned to the gold and, in addition, the pyrite contained in the residues can be converted into

* Under current regulations, the A.E.C. will purchase domestic ores having a uranium content down to 0.1 per cent U_3O_8.

sulfuric acid for use in processing; the gangue consists mainly of inert quartz. Uranium production from the gold-ore residues has already been started and it is expected to provide important amounts of the element.

8.8. Low-grade potential sources within the United States, containing 0.01 per cent or less of uranium, include the extensive phosphate fields of Florida and Idaho, the Tennessee oil shales, and the uraniferous lignites of Wyoming and the Dakotas. From the phosphates, a small amount of uranium is being recovered in the production of phosphoric acid, and processes are being developed for its extraction as a by-product in fertilizer manufacture. The recovery of uranium from lignite and shales does not appear to be economic at present. Nevertheless, from the long-range standpoint, the large amounts of uranium in these low-grade sources cannot be overlooked, and possible methods of uranium extraction are being studied.

8.9. In conclusion, it should be noted that very large quantities of uranium are present in sea water, but at the extremely low concentration of 1 to 2 ppb. Practicable recovery of materials at this concentration is beyond the reach of any presently known processing method.

Treatment of Uranium Ores*

8.10. The higher grade pitchblende ores can be concentrated ("upgraded") to about 50 per cent U_3O_8 at the mine by physical methods, e.g., crushing, screening, washing, flotation, and gravity separation, before being shipped to a processor (in the United States) for further purification of the uranium. The tailings from the physical concentration step and the lower grade pitchblende ores are upgraded by chemical methods. The uranium is extracted from the ore, along with other metals, by leaching with dilute sulfuric acid in the presence of an oxidizing agent, e.g., sodium chlorate or manganese dioxide. After decantation and filtration, the uranium may be removed from some of the liquors by adding magnesium oxide at controlled pH to precipitate a crude form of magnesium uranate, known as MgX. This has a sufficiently high uranium content to permit of its use in the refining operation.

8.11. Many of the carnotite ores from the Colorado Plateau contain vanadium in sufficient amount for commercial extraction to be worth while. As indicated above, in the past these deposits were processed for vanadium as the sole product, but now the uranium is also recovered. The procedure is determined by the nature of the ore and gangue, and many minerals have been treated either by the "acid" or the "carbonate" process as described below.

8.12. In the *acid process*† the carnotite ore is crushed, mixed with sodium chloride, and roasted. The hydrogen chloride gas evolved is absorbed in water to form hydrochloric acid for use in a later stage of the operation. The calcined ore is leached with water, and this removes about 75 per cent of the vanadium

* R. D. Macdonald, *loc. cit.*; W. L. Lennemann, *Eng. Mining J.*, **155**, No. 9, 104 (1954).
† V. L. Saine and K. B. Brown, Report AECD-3241.

as soluble sodium vanadate. The residue is then treated with a mixture of hydrochloric and sulfuric acids which extracts about 90 per cent of the uranium and an additional 15 per cent of the vanadium. The acid leach is reduced with iron, and ammonia is added to precipitate the uranium (and other metals, including the iron) as "green sludge." The sludge is dissolved in sulfuric acid and the solution oxidized with sodium chlorate; the pH is then adjusted to 3, at which point the vanadium is precipitated as iron vanadate. The residual solution from this step, containing nearly all the uranium, is treated for the removal of alumina and silica, and finally sodium diuranate or "soda salt,"

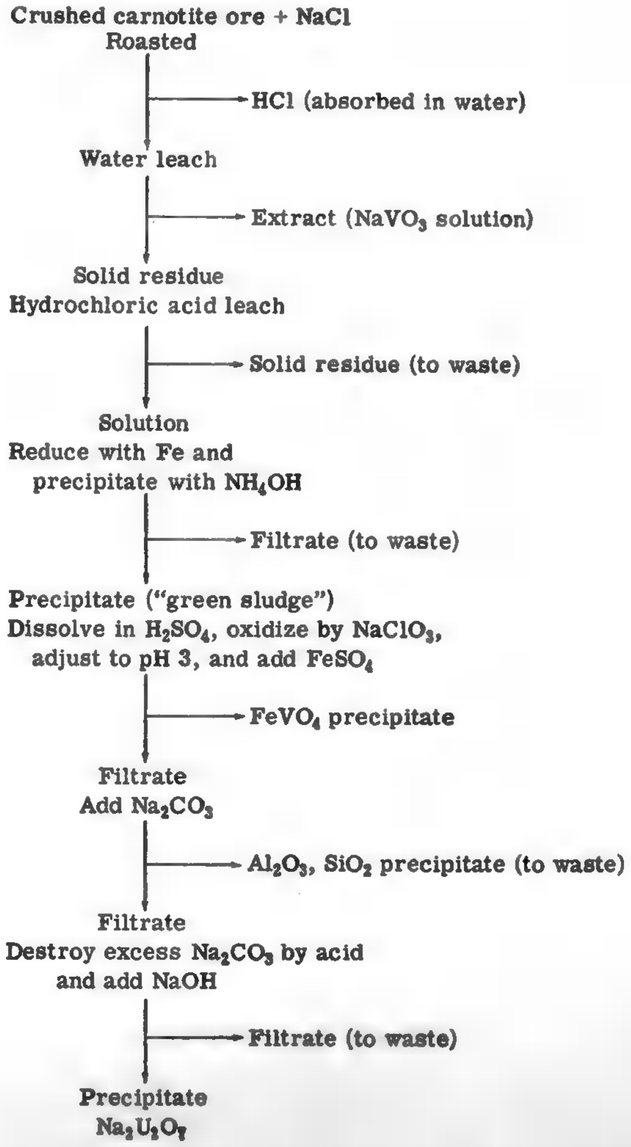


FIG. 8.1. Simplified flow sheet of acid process for uranium extraction

approximately $Na_2U_2O_7$, is precipitated by means of sodium hydroxide. A simplified flow sheet of the acid process is shown in Fig. 8.1.

8.13. The first stage of the *carbonate process** is similar to that of the acid process, i.e., roasting of the crushed carnotite ore with sodium chloride. The roasted ore is quenched in sodium carbonate solution, and most of the uranium and vanadium is thereby extracted. Upon neutralizing (pH 6.5 to 7) with sulfuric acid, the uranium is precipitated as sodium uranyl vanadate, the excess vanadium remaining in solution as sodium vanadate.† The precipitate is heated

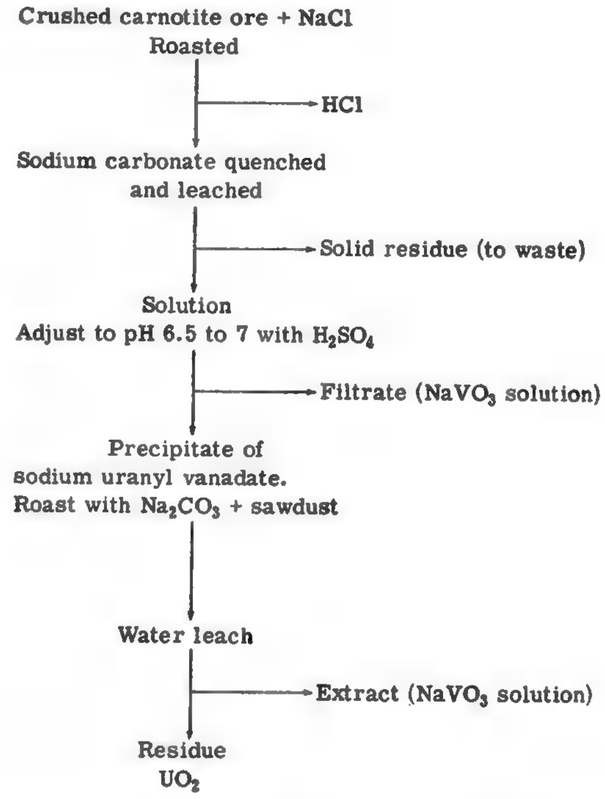


FIG. 8.2. Simplified flow sheet of carbonate process for uranium extraction

with a mixture of sodium carbonate, sodium chloride, and sawdust and then leached with water. The vanadium is extracted as sodium vanadate, and the uranium remains in the residue as uranium dioxide (UO_2). In Fig. 8.2 is given a simplified flow sheet of the carbonate process.

8.14. Ores containing large proportions of calcium carbonate cannot be treated by either of the processes described above. In such cases different procedures must be used, e.g., direct extraction of uranium from the ore into sodium car-

* E. L. Leyshon and C. F. Coleman, Report AECD-3223.

† The precipitate is essentially a synthetic (sodium) carnotite, having the approximate composition $Na_2O \cdot 2UO_3 \cdot V_2O_5 \cdot xH_2O$ or $Na_2(UO_2)_2(VO_4)_2 \cdot xH_2O$.

bonate solution. The same is true for ores containing little or no vanadium, e.g., the carnotite from Grants, New Mexico, and the uraninite-autunite deposits from Marysvale, Utah. In the latter case the ore is first roasted to destroy carbonaceous matter and improve its filtration characteristics, and then leached with dilute sulfuric acid. The extract is reduced with iron or aluminum, and addition of ammonia and sodium phosphate at pH 2.6 results in the precipitation of uranium phosphate. This is heated with sodium carbonate together with sodium chloride as flux; the soluble material is leached out with water leaving uranium dioxide as the residue.

8.15. A method for the extraction of uranium from the South African (Rand) gold-ore residues (§ 8.7) involves separation of the pyrite by flotation. The pyrite is roasted in air and the sulfur dioxide produced is converted into sulfuric acid; this is used to leach the uranium from the gold-ore residues. From the resulting liquors the uranium can be extracted by an ion-exchange process and the product is finally calcined to yield "black oxide," crude U_3O_8, which is shipped to the United States.

8.16. Although considerable quantities of phosphatic minerals containing uranium are processed to yield fertilizer and other phosphorus products, it is generally difficult to recover the uranium without introducing extensive, and often impractical, changes into the phosphate industry. However, in certain cases the process is such that uranium extraction is possible without disturbing the main procedure. One of the most important instances is the recovery of the element from industrial phosphoric acid, which is produced as an intermediate in the manufacture of "triple phosphate."

8.17. The so-called "leached zone," which is part of the overburden of the Florida phosphate pebble beds, contains significant quantities (0.01 to 0.015 per cent) of uranium. The material, mainly a mixture of silica, clay, and aluminum and calcium phosphates, has hitherto been discarded by throwing it back into the mined-out pits. However, processes are now being developed for recovering the uranium from the leached zone with phosphate fertilizer and possibly aluminum oxide as by-products.

8.18. It may be mentioned that experimental studies have been made of the recovery of uranium from oil shales and, to a lesser extent, from lignites. Since these sources are unlikely to be economically significant for many years, by which time entirely different extraction processes may be developed, the subject need not be considered further here. Reference to the recovery of uranium from monazite sand will be made later (§ 8.31).

Purification of Uranium from Concentrates

8.19. Because the uranium for use as a reactor fuel must be especially free from elements having appreciable neutron-capture cross sections, the products of the processes already described are subjected to further purification. For this purpose solvent extraction has been used for some time. Until recently diethyl ether was employed exclusively as the solvent,* but this is being replaced by other organic media, of the type referred to in § 7.164, e.g., *n*-tributyl phosphate, which are less volatile and less inflammable.

8.20. In the diethyl ether process, the concentrate, consisting of uranium oxide (UO_2 or U_3O_8), sodium diuranate, MgX, or other product, is treated with nitric acid to form uranyl nitrate solution. Lead and radium, if present, are removed from the solution by precipitation as sulfate with barium sulfate as carrier. The filtered uranyl nitrate solution is then extracted with diethyl ether. The organic phase containing the uranium is scrubbed with dilute nitric acid for further purification, and the uranyl nitrate is finally back-extracted into the aqueous phase with water. The solution is evaporated to remove nitric acid and water, and the final product is usually sufficiently pure for reactor applications.

8.21. In a modified form of the solvent extraction procedure used in the United States, the uranyl nitrate is extracted directly from a slurry by means of an organic solvent, without the necessity for filtration. A nitric acid digest of the finely ground uranium concentrate is fed into the middle of an extraction column. As it runs down the slurry meets an upflowing stream of organic solvent which removes the uranyl nitrate. In the upper half of the column the organic phase is scrubbed with nitric acid, and the uranyl nitrate is then back-extracted with demineralized water in a stripping column (cf. § 7.174).

Preparation of Uranium Metal and Hexafluoride†

8.22. For use in nuclear energy work, uranium is generally required either in the form of uranium metal, for fabrication into fuel elements, or as hexafluoride ("hex") to constitute the feed for the separation of the isotopes by the gaseous-diffusion method (§ 8.60). In each case uranium tetrafluoride (UF_4), sometimes referred to as "green salt," must first be prepared and this can be converted either into metal or into hexafluoride.

8.23. Several different methods have been used for the production of uranium tetrafluoride, but all involve the intermediate formation of the dioxide ("brown oxide"). The concentrated solution of purified uranyl nitrate may be heated directly in a furnace to yield the trioxide ("orange oxide"), i.e., UO_3, which is subsequently reduced with hydrogen at about 600°C (1100°F) to the dioxide. Alternatively, the uranyl nitrate solution is treated either with ammonia (to precipitate ammonium diuranate) or with hydrogen peroxide (to precipitate hydrated uranium peroxide, $UO_4 \cdot xH_2O$). The precipitate is filtered off, dried,

* See, for example, H. D. Smyth, "Atomic Energy for Military Purposes," U. S. Government Printing Office, 1945, § 6.12; K. E. B. Jay, "Britain's Atomic Factories," H. M. Stationery Office, 1954, p. 16.

† C. Eichner, B. Goldschmidt, and P. Vertes, *Bull. Soc. Chim.*, 140 (1951), Report NYO-1340 (translation); J. Van Impe, *Chem. Eng. Prog.*, 50, 230 (1954); H. M. Finniston, *Research*, 5, 456 (1952); K. E. B. Jay, *op. cit.*, p. 17; see also, A. S. Newton, U. S. P., 2,446,780 (1948).

and heated to form uranium trioxide, in each case; this oxide is then reduced to the dioxide with hydrogen, as stated above.

8.24. The simplest procedure for converting uranium dioxide to the tetrafluoride is to heat the oxide in a furnace and pass anhydrous hydrogen fluoride gas over it, when the reaction

$$UO_2 + 4HF \rightarrow 2H_2O + UF_4$$

takes place. The resulting "green salt" is a solid having a high melting point (about 960°C or 1760°F) and so is left behind. Because of the highly corrosive nature of hydrogen fluoride, special alloys are used for the furnace lining and for the pipes through which the gas passes.

8.25. For production of uranium tetrafluoride on a moderate scale two other methods have been employed that do not require gaseous hydrogen fluoride. In one a 40 per cent solution of aqueous hydrofluoric acid is allowed to react with uranium dioxide in a hard-rubber vat. The dioxide is thereby converted into solid uranium tetrafluoride which is filtered, washed, and carefully dried by heating. In the other method the double salt $NH_4F \cdot UF_4$ is first formed by the action of solid ammonium acid fluoride (NH_4HF_2) on uranium dioxide at 150°C (300°F). The double salt is then slowly heated in a vacuum furnace, up to about 500°C (930°F), when the ammonium fluoride distills off, leaving the uranium tetrafluoride.

8.26. In the conversion into uranium metal, the finely powdered tetrafluoride is heated in a steel mold or bomb with either calcium or magnesium of high purity; the reaction taking place, e.g., with calcium, is then

$$UF_4 + 2Ca \rightarrow U + 2CaF_2.$$

The calcium (or magnesium) fluoride forms a slag on the surface of the uranium, from which it can be separated. In order to prevent entry of impurities into the uranium metal, the reaction vessel is lined with a refractory material, such as calcium fluoride (fluorite). The massive metal is then removed, melted (at 1300°C or 2370°F) in a graphite crucible under vacuum to remove volatile impurities, and poured into molds. The ingots so produced are suitable for various types of fabrication to be described later (§ 8.47). Some indication of the purity of the product is given by the results in Table 8.1, obtained mainly by spectrographic analysis.* In addition to the elements mentioned in the table, tests were made for the following which were either absent entirely or present in undetectable amounts: arsenic, barium, bismuth, cadmium, cobalt, cesium, germanium, indium, rubidium, antimony, tin, columbium (niobium), gallium, hafnium, palladium, tantalum, titanium, vanadium, and zirconium.

TABLE 8.1. TYPICAL ANALYSIS OF URANIUM METAL FOR REACTOR FUEL

Element	*Ppm*	*Element*	*Ppm*
Carbon	40	Chromium	<5
Iron	28	Magnesium	<5
Aluminum	<20	Nickel	<5
Oxygen	16	Copper	3
Silicon	10	Molybdenum	<1
Potassium	<10	Lead	<1
Manganese	8	Silver	≤0.5
Nitrogen	7	Boron	<0.2

8.27. If the uranium tetrafluoride is to be converted to the hexafluoride, the former is allowed to react with fluorine gas at temperatures somewhat above

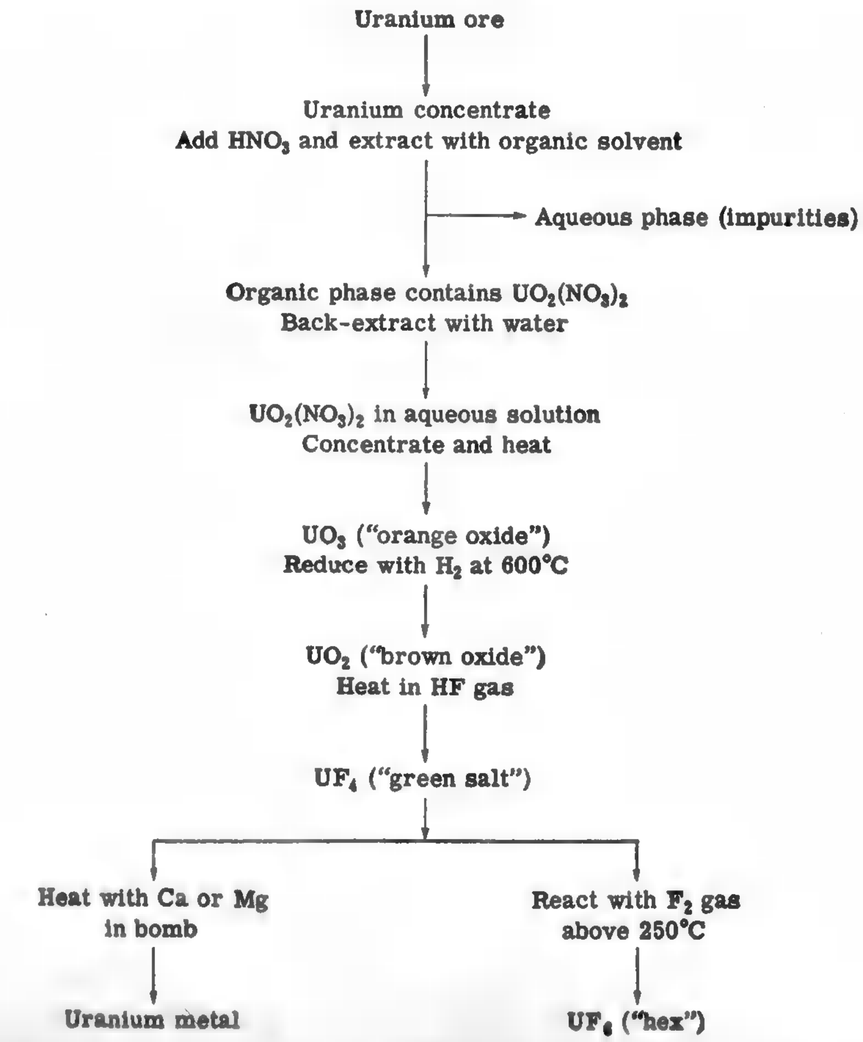


FIG. 8.3. Summary flow diagram for uranium metal and uranium hexafluoride production

* J. Van Impe, *loc. cit.*

250°C (480°F);* the reaction occurring is simply $UF_4 + F_2 \rightarrow UF_6$. Since uranium hexafluoride, in contrast to the tetrafluoride, is very volatile,† it is vaporized as fast as it is formed and is collected (as solid) in traps cooled to temperatures well below 0°C (32°F). A general flow sheet showing the various stages involved in the production of either uranium metal or uranium hexafluoride from the ore is given in Fig. 8.3.

Thorium Sources

8.28. The average abundance of thorium in the earth's crust has been estimated to be three times as great as that of uranium, i.e., about 12 ppm. However, there are so few recognized deposits containing significant quantities of thorium that the total amount which can be extracted at a reasonable cost may be less than that of uranium. The richest naturally occurring thorium minerals are *thorite* and *orangite*, both silicates containing 50 to 70 per cent of thorium dioxide (ThO_2), but the known deposits are too small to provide an important source of the element. Most commercial supplies are obtained from the monazite-bearing sands, of which the largest amounts are found in the state of Travancore, India. There are also considerable quantities of monazite sand in Brazil; lesser amounts are found in other parts of the world, including North and South Carolina, Florida, and Idaho in the United States.‡

8.29. Monazite is essentially a mixture of rare-earth phosphates, containing a small percentage of thorium and a still smaller proportion of uranium. The alluvial deposits, resulting from the weathering of granites, include a great deal of silica (sand), and these are treated by standard physical and mechanical methods to concentrate the monazite. The resulting product may contain from 5 to 8 per cent of thorium, 0.15 to 0.25 per cent of uranium, and some 50 per cent of rare-earth elements, mostly in the form of phosphates. There are also variable amounts of silica as well as of iron, titanium, and other metals.

Extraction of Thorium

8.30. In the past the thorium—and a small proportion of cerium—was extracted from monazite for the manufacture of Welsbach incandescent mantles, but at the present time recovery of the uranium, and also of the rare-earth elements, is considered desirable. Two processes have been developed for achieving these ends; they are distinguished by the fact that they involve alkaline or acid digestion, respectively.

* J. J. Katz and E. Rabinowitch, *op. cit.*, p. 398.

† Pure uranium hexafluoride, which is a solid at ordinary temperatures, sublimes at 56.5°C (133.7°F); that is to say, it then has a vapor pressure of 1 atm (760 mm Hg). The pure liquid can exist only under pressures in excess of 1134 mm and temperatures above 64.05°C (147.3°F), the triple point.

‡ The Indian reserves are estimated to contain about 170,000 tons of thorium and the Brazilian reserves about 10,000 tons (U. S. Bureau of Mines, "Minerals Year Book," U. S. Government Printing Office, 1949).

8.31. The first stage in the *alkaline process** is digestion of the monazite concentrate with 73 per cent sodium hydroxide solution at about 140°C (284°F). This results in the conversion of the phosphate into soluble sodium triphosphate, while the thorium, uranium, and rare-earth elements are left as insoluble hydrous oxides. The latter are filtered off and dissolved in hot concentrated hydrochloric acid, and then by the addition of alkali the pH is adjusted to 5.8. A hydrous

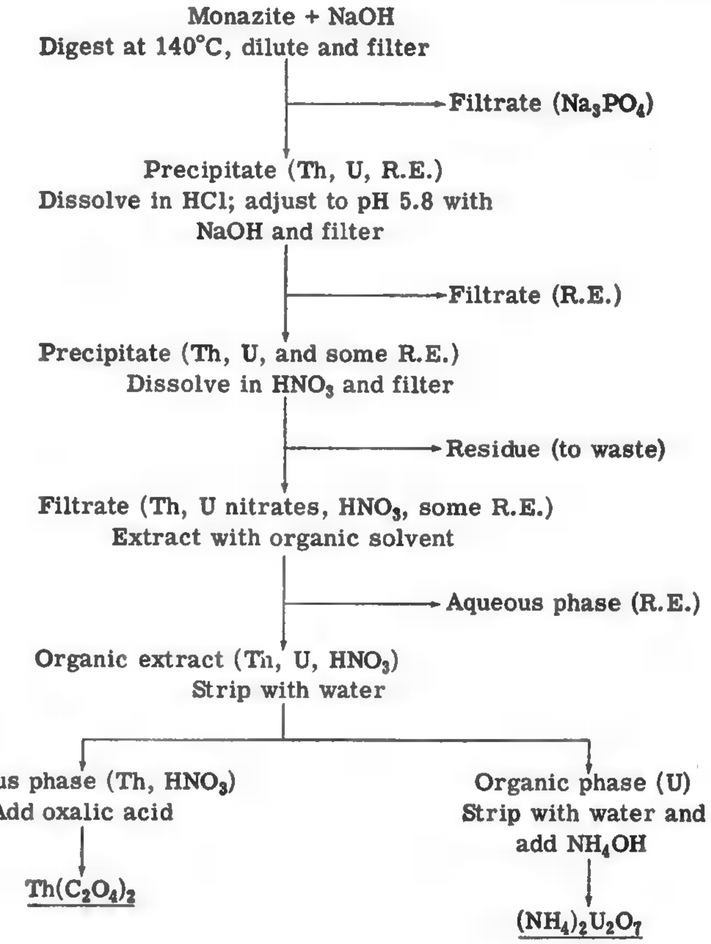


Fig. 8.4. Simplified flow sheet of alkaline process for treatment of monazite sand

oxide precipitate is thus obtained containing over 99 per cent of the thorium and uranium with only 2 to 3 per cent of the rare earths, the greater proportion of the latter remaining in solution. Further purification of the thorium and uranium and separation from each other are achieved by solvent extraction of the nitrate solution obtained by dissolving the hydrous oxide precipitate in nitric acid. A flow sheet of the process is outlined in Fig. 8.4.

* A. E. Bourne, G. D. Calkins, J. W. Clegg, and R. B. Filbert, Jr., *Chem. Eng. Prog.*, **50**, 235 (1954).

8.32. In the *acid digestion process** the monazite is treated with hot concentrated (93 per cent) sulfuric acid, so that the thorium, uranium, and rare-earth elements pass into solution as sulfates together with phosphoric acid. Upon dilution of the solution and adjustment of the pH to about 1.0 with ammonium hydroxide, essentially all the thorium is precipitated accompanied by about 5 per cent of the rare earths. Because of the great preponderance of the latter in the monazite sand, the precipitate consists of about half thorium phosphate and half occluded rare-earth sulfates. The uranium and the bulk of the rare earths remain in solution, but upon raising the pH to 2.3, by the addition of more ammonia, most of the rare earths are precipitated. Finally, by adjusting the pH to about 6.0, the precipitate formed contains nearly all of the uranium

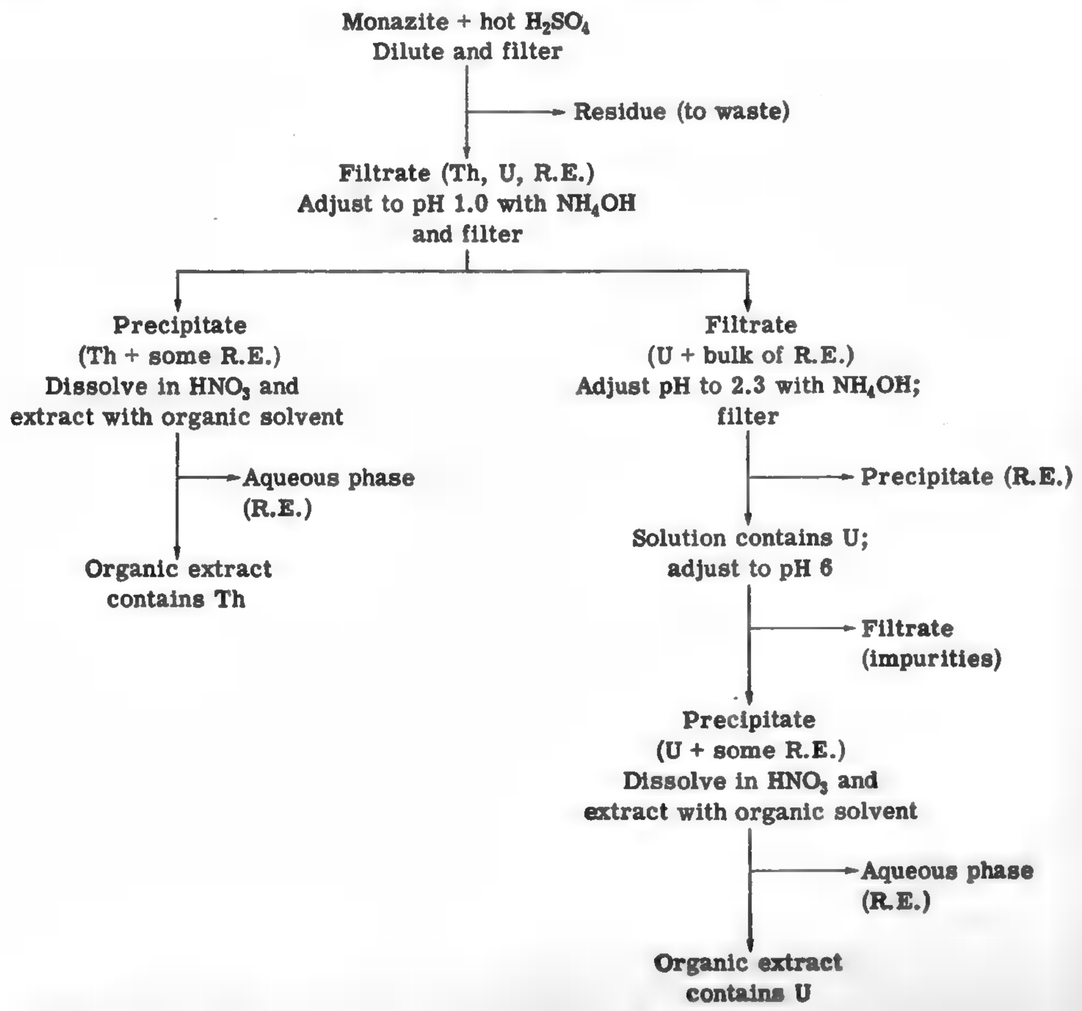


FIG. 8.5. Simplified flow sheet of acid process for treatment of monazite sand

* M. Smutz, G. L. Bridger, K. G. Shaw, and M. E. Whatley, *Chem. Eng. Prog. Symposium Series*, No. 13, **50**, 167 (1954).

together with the residual rare earths. The thorium and uranium precipitates are purified from the accompanying rare-earth elements by solvent extraction after dissolving in nitric acid. A simplified flow sheet of the acid digestion process is given in Fig. 8.5.

PRODUCTION OF THORIUM METAL*

8.33. The problem of reducing thorium compounds to the metal is somewhat analogous to that encountered with uranium, and similar methods have been employed to obtain the two metals. Thus metallic thorium can be prepared by reduction in a bomb of the tetrachloride or tetrafluoride with either sodium, calcium, or magnesium. Thorium oxalate is first heated in air to 650°C (1200°F) to form thorium dioxide (ThO_2), and then hydrogen fluoride gas is passed over the dioxide, heated to 550°C (1020°F), to yield thorium tetrafluoride (ThF_4), as described for uranium (§ 8.24).

8.34. The reduction of thorium tetrafluoride to the metal by calcium or magnesium is rendered difficult because of the high melting point of thorium (about 1690°C or 3075°F). One way in which this difficulty may be overcome is to introduce zinc into the reduction system, since this forms a relatively low melting point alloy with thorium. When the reduction is complete, the zinc can be removed by heating the alloy in a vacuum. The zinc distils off, leaving the thorium either as a spongy mass or in the form of liquid if the temperature is high enough.

8.35. Thorium metal of very high (>99.9 per cent) purity can be obtained by the deBoer iodide process. Thorium tetraiodide, produced by the action of iodine vapor on machined chips of the impure metal, is thermally decomposed in a sealed Vycor tube by means of an electrically heated wire filament. The pure thorium deposits on the filament in a loosely knit crystalline form which can be converted into an ingot by arc melting. The iodine liberated in the decomposition is available for reaction with more impure thorium, so that the process is essentially continuous.

PROPERTIES OF URANIUM METAL†

GENERAL AND PHYSICAL PROPERTIES

8.36. Massive uranium in the pure state is a dense, hard, white to grayish-white metal with a distinct luster when freshly prepared. It tarnishes readily in air due to the formation of a loosely adherent oxide film. Because of this rapid reaction with oxygen, the finely divided metal is pyrophoric, and fine chips

* H. M. Finniston, *loc. cit.*; J. R. Keeler, *Chem. Eng. Prog. Symposium Series*, No. 11, **50**, 57 (1954).

† For further details and references, see J. J. Katz and E. Rabinowitch, *op. cit.*, Chapters 5 and 6; H. A. Saller and F. A. Rough, *Chem. Eng. Prog. Symposium Series*, No. 11, **50**, 63 (1954); H. M. Finniston, *loc. cit.*; *Times Science Review* (London), No. 6, 11 (1952); C. E. Hampel (Ed.), "Rare Metals Handbook," Reinhold Publishing Corp., 1954, Chapter 26 (by G. Meister).

or turnings are a serious fire hazard if exposed to air. Treatment of uranium metal at elevated temperatures must therefore be carried out in an inert atmosphere or in a vacuum. Fortunately the vapor pressure of both the solid and the liquid for an appreciable temperature range above the melting point is relatively low, so that a high vacuum can be used for casting or for removal of more volatile impurities (cf. § 8.26).

8.37. Between ordinary atmospheric temperature and its melting point (1133°C or 2070°F), uranium exists in three different crystalline (allotropic) forms. This fact has a significant influence on the treatment and behavior of reactor fuel elements at elevated temperatures. The three forms are called the alpha, beta, and gamma phases, respectively, and their characteristic properties are indicated in Table 8.2.* The cell dimensions and the density, calculated from X-ray measurements, are for the stated temperatures.† Owing to the presence of small amounts of carbon, the actual density of massive (alpha) uranium is usually less than that given in the table.

TABLE 8.2. PROPERTIES OF THE CRYSTALLINE FORMS OF URANIUM

Property	Alpha Phase	Beta Phase	Gamma Phase
Stability range, °C	Below 662	662 to 772	772 to 1133 (m.p.)
°F	Below 1224	1224 to 1422	1422 to 2070
Crystalline form	Orthorhombic	Tetragonal	Body-centered cubic
Cell dimensions, A	(20°C)	(720°C)	(805°C)
a_0	2.858	10.76	3.524
b_0	5.877	—	—
c_0	4.955	5.656	—
Density (calc.), g/cm³	19.00	18.11	18.06
General characteristics	Soft and ductile	Hard and brittle	Very soft

8.38. The structures of the alpha and gamma phases are relatively simple. The alpha phase may be regarded as having a distorted hexagonal close-packed structure, whereas the gamma phase is body-centered cubic. The beta phase is somewhat more complex, the tetragonal unit cell containing 30 atoms.

8.39. The low symmetry of the orthorhombic system leads to considerable anisotropy in the alpha phase of uranium, as is indicated by the cell dimensions in Table 8.2. One consequence is that the thermal expansion is anisotropic, as may be seen from the approximate average coefficients of thermal expansion in the principal crystallographic directions recorded in Table 8.3; the temperature range is from 25 to 650°C (77 to 1200°F). Thus, while the dimensions parallel to the [100] and [001] directions increase, although to different extents, with

* Cf. J. Thewlis, *Nature*, **168**, 198 (1951); *Acta Cryst.*, **5**, 790 (1952); C. W. Tucker, Jr. and P. Senio, *ibid.*, **6**, 753 (1953).

† The estimated values for the beta phase at ordinary temperatures are $a_0 = 10.59$ A, $c_0 = 5.634$ A; density = 18.56 g/cm³.

increasing temperature, there is a decrease parallel to the [010] plane direction. Upon heating or cooling, therefore, crystals of the alpha phase of uranium undergo considerable distortion.

TABLE 8.3. COEFFICIENTS OF THERMAL EXPANSION OF ALPHA PHASE OF URANIUM FROM 25 TO 650°C (77 TO 1200°F)

Direction	*Average linear coefficient, per °C*
a [100]	36.7×10^{-6}
b [010]	-9.3×10^{-6}
c [001]	34.2×10^{-6}

8.40. The structure of the alpha phase is not at all typical of a metal but resembles more that of the pseudo-metals, such as tellurium. This is manifested in the relatively low thermal and electrical conductivity of alpha uranium. The thermal conductivity is about 0.060 cal/(sec)(cm²)(°C/cm)*, and the electrical resistivity is 25 to 30 $\times 10^{-6}$ ohm-cm at 25°C (78°F).† The values are undoubtedly dependent on the crystallographic direction, and the results quoted are to be regarded as approximate averages. The close-packed cubic structure of the gamma phase is characteristic of many metals, and it is of interest to record that the transition from beta to gamma (and also from alpha to beta) is accompanied by a decrease in electrical resistivity; there is probably an accompanying increase in thermal conductivity.

8.41. Although the phase changes take place fairly sharply at the transition temperatures, i.e., 662°C (1224°F) for $\alpha \rightleftharpoons \beta$ and 772°C (1422°F) for $\beta \rightleftharpoons \gamma$, it is possible by suitable alloying to retain the beta and gamma structures at room temperatures. For example, the presence of a relatively small amount of chromium results in a stabilization of the beta phase, whereas certain solid solutions of uranium and molybdenum retain the gamma structure.

Mechanical Properties

Hardness

8.42. The hardness of uranium is dependent on the treatment of the metal. The average hardness, at ordinary temperatures, of cast annealed metal is in the region of 200 to 220 on the Brinell scale or about 90 on the Rockwell B scale. As a result of cold-working, the hardness can be increased from Rockwell B 90 to 115 or more. Most of this increase occurs during the first 20 per cent reduction in thickness, further cold-work having little effect. The hardness of cold-worked uranium begins to decrease upon heating to 150°C (300°F), and

* To convert thermal conductivities in metric units to values in engineering units, i.e., Btu/(hr)(ft²)(°F/ft), multiply by 242.

† For purposes of comparison, it may be noted that, for copper at ordinary temperatures, the thermal conductivity is about 1 cal/(sec)(cm²)(°C/cm) and the electrical resistivity is 1.72×10^{-6} ohm-cm.

complete annealing occurs at 600 to 700°C (1110 to 1290°F). Increase of temperature is accompanied by a decrease in hardness of the alpha phase. By 650°C (1200°F), the Brinell number has dropped to 13, but there is an increase to 30 or 40 Brinell units when the beta phase is formed. Further increase of temperature results in a continued decrease in hardness, and the gamma phase above 772°C (1422°F) is so soft that it cannot be measured on the Brinell scale.

Elasticity

8.43. The proportionality between stress and strain for the alpha phase occurs only at loads of less than 10 to 15 $\times$ 10^3 psi. The modulus of elasticity is then 15 to 25 $\times$ 10^6 psi. The range in values is probably related to the anisotropic nature of the alpha form. With increasing temperature both the proportional limit and the elasticity decrease rapidly. Because the proportional limit for uranium is so relatively low, the metal has been described as "semiplastic." In accordance with this semiplastic nature, the Poisson's ratio of uranium is low. The observed values depend on the treatment of the material, but most reported data are in the range from 0.2 to 0.25.

8.44. Since stress-strain curves for uranium show a very low proportional limit, the yield strength is usually designated as the stress required to produce a strain 0.2 per cent larger than that which would be expected from elastic behavior. This value varies from 25 to 130 $\times$ 10^3 psi, depending on prior cold work, impurity content, method of fabrication, and orientation of the test sample. Fabricated uranium is highly anisotropic in its properties and may show an appreciable difference in yield strengths measured in the direction of working or perpendicular to this direction.

Strength

8.45. At ordinary temperatures the ultimate tensile strength of alpha uranium varies between 50 and 200 $\times$ 10^3 psi; the highest values result from cold-working and the lowest from annealing at high temperature. The tensile strength is somewhat improved by quenching; thus, rolled, forged, or swaged specimens quenched from 600 to 1000°C (1110 to 1830°F) have tensile strengths of 90 to 130 $\times$ 10^3 psi. With increase of temperature the tensile strength decreases rapidly; one specimen, for example, decreased from 53 $\times$ 10^3 psi at room temperature to 27 $\times$ 10^3 psi at 150°C (300°F) and 13 $\times$ 10^3 psi at 600°C (1110°F).

8.46. It will be evident from the foregoing remarks that the mechanical properties of metallic uranium vary considerably with the condition and previous treatment of the specimen. The summarized data in Table 8.4 may be regarded, however, as being roughly applicable to annealed (alpha phase) uranium at ordinary temperatures.

TABLE 8.4. MECHANICAL PROPERTIES OF ANNEALED URANIUM

Tensile strength, psi	50 to 200 $\times$ 10^3
Yield strength (0.2% offset), psi	25 to 130 $\times$ 10^3
Modulus of elasticity, psi	15 to 25 $\times$ 10^6
Proportional limit, psi	10 to 15 $\times$ 10^3
Poisson's ratio	0.2 to 0.25

Fabrication of Uranium Metal

8.47. Uranium can be fabricated by conventional means, including casting, rolling, extrusion, forging, swaging, drawing, and machining. Hot-rolling of the alpha phase is a useful method for forming the metal. Because of the ease with which uranium oxidizes, especially at higher temperatures, it must be protected from air during fabrication, either by means of a fused salt or by an atmosphere of inert gas. Although the metal can be machined moderately easily, suitable lubricants and coolants are required to prevent oxidation.

8.48. Uranium parts can be joined by welding or brazing. Fusion welding is achieved by using a Heliarc torch in an inert atmosphere. Brazing is rendered somewhat difficult by the formation of brittle alloys between the uranium and most of the common brazing alloys. Satisfactory results have been obtained by first plating the uranium with nickel or silver and then brazing the plated metal by conventional methods.

Uranium Alloys

8.49. Because of its low strength and poor corrosion resistance, as well as for other reasons, investigations have been made of combinations of uranium with various metals in the hope of finding alloys which do not have the drawbacks of uranium itself. The greatest improvements in both mechanical properties and in corrosion resistance have been observed with alloys containing small amounts of the added metal in solid solution. Among the elements having appreciable solubility in the solid uranium phase at elevated temperatures mention may be made of zirconium, titanium, columbium, and molybdenum. A number of elements form intermetallic compounds with uranium. One of these is aluminum, which has been extensively used to alloy with uranium for the fabrication of reactor fuel elements. Three compounds, namely, UAl_2, UAl_3, and UAl_4, have been identified.*

PROPERTIES OF THORIUM METAL†

General and Physical Properties

8.50. Although thorium is a dense metal, it is less dense, as well as softer, than uranium. Freshly prepared surfaces have a bright metallic silvery luster, which soon darkens upon exposure to the atmosphere, due to the formation of an oxide

* P. Gordon and A. R. Kaufmann, Report AECD-2083.

† H. A. Saller, *Proc. Conf. on Nuc. Eng.*, Berkeley, 1953, p. 11; *Science*, **119**, 4 (1954); J. R. Keeler, *Chem. Eng. Prog. Symposium Series*, No. 11, **50**, 57 (1954); C. A. Hampel (Ed.), "Rare Metals Handbook," Reinhold Publishing Corp., 1954, Chapter 23 (by W. C. Liliendahl).

layer. Finely divided thorium is pyrophoric, so that care must be maintained when castings are machined to avoid combustion of the chips. The massive metal scales rapidly in air, especially at elevated temperatures, and also in boiling water.

8.51. Thorium melts at 1690°C (3075°F), and over most of the range from room temperature to the melting point it has a face-centered cubic structure. Between 1400 and 1500°C (2550 and 2730°F) a phase change, leading to a body-centered cubic form, has been reported. The fact that thorium belongs to the cubic system means that its properties will be isotropic; in this respect, therefore, it has advantages over uranium which exhibits anisotropic behavior over a considerable temperature range.

8.52. The edge length of the face-centered cubic cell at ordinary temperatures is 5.08 A, and the corresponding calculated density of thorium is 11.7 g/cm³. The experimentally determined value for the arc-melted metal is 11.66 g/cm³. The linear coefficient of thermal expansion is 12×10^{-6} per °C between 25 and 500°C (78 and 930°F).

8.53. The thermal conductivity of thorium is 0.090 at 100°C (212°F) and increases to 0.108 cal/(sec)(cm²)(°C/cm) at 650°C (1200°F). Its electrical resistivity is about half that of alpha uranium, namely, 18×10^{-6} ohm-cm. This is in accord with the more pronounced metallic character of the face-centered cubic structure. Some of the mechanical properties of annealed thorium metal at room temperatures are recorded in Table 8.5. By cold-rolling, the tensile strength can be increased to 60×10^3 psi, and the hardness is appreciably increased at the same time.

TABLE 8.5. MECHANICAL PROPERTIES OF ANNEALED THORIUM

Tensile strength, psi	32 to 37×10^3
Yield strength (0.2% offset), psi	24 to 32×10^3
Modulus of elasticity, psi	10×10^6
Proportional limit, psi	22×10^3
Shear modulus, psi	4×10^6

Fabrication of Thorium Metal

8.54. Like uranium, metallic thorium can be fabricated by a variety of standard methods. The material is very ductile and considerable reduction is possible before annealing is necessary. It can be forged, extruded, hot- and cold-rolled, swaged, and drawn. Although the metal is semi-brittle under impact at ordinary temperatures, it becomes quite tough at about 250°C (480°F). The machining characteristics of thorium are similar to those of mild steel. In order to prevent oxidation of the metal during fabrication, appropriate steps must be taken to prevent access of air.

8.55. Joining of thorium either to itself or to other metals has proved difficult.

Various attempts at welding and brazing have not been very successful, for one reason or another.

Thorium Alloys

8.56. The investigation of thorium alloys has had two important objectives: the discovery of materials with improved mechanical properties and with greater corrosion resistance. Thorium forms intermetallic compounds with many other metals and several of these alloys are appreciably stronger than thorium itself. The presence of small proportions of carbon produces a marked increase in the hardness of the metal. Although thorium is readily attacked by boiling water, as already indicated, an alloy containing 25 to 30 per cent by weight of zirconium is reported to have shown no change in weight after 376 hours in boiling water.

PROPERTIES OF PLUTONIUM METAL*

General and Physical Properties

8.57. Because of its special importance in atomic weapons, relatively little information has been published on the physical and metallurgical properties of plutonium. Nevertheless, since plutonium-239 has interesting nuclear properties, it may one day play an important role as the fuel material in breeder reactors (§ 1.147). The metal has been obtained by reduction of the fluoride, either PuF_3 or PuF_4, with one of the alkaline-earth elements, e.g., barium in the vapor form.

TABLE 8.6. PROPERTIES OF SOLID PHASES OF PLUTONIUM

Phase	Stability Range	Structure	Density (g/cm³)	Thermal Expansion Coeff. per °C × 10⁶
α	Below 122°C (252°F)	Complex	19.7 (25°C)	51
β	122 to 206°C (252 to 403°F)	Complex	17.6 (150°C)	38
γ	206 to 319°C (403 to 607°F)	Face-centered orthorhombic	17.2 (210°C)	35
δ	319 to 451°C (607 to 844°F)	Face-centered cubic	15.9 (320°C)	−10
δ'	451 to 476°C (844 to 889°F)	Face-centered tetragonal	16.0 (465°C)	−120
ϵ	476 to 640°C (m.p.) (889 to 1185°F)	Body-centered cubic	16.5 (500°C)	26

* E. R. Jette, *J. Chem. Phys.*, **23**, 365 (1955); see also, C. S. Smith, *Metal Prog.*, **65**, No. 5, 81 (1954); *Phys. Rev.*, **94**, 1068 (1954); W. B. H. Lord, *Nature*, **173**, 534 (1954); J. G. Ball, *et al.*, *ibid.*, **173**, 535 (1954).

8.58. Plutonium is a heavy metal, its density being near to that of uranium. Fresh surfaces have a distinctly metallic luster, but they tarnish readily upon exposure to air. The physical metallurgy of plutonium is complicated by the fact that six allotropic forms, designated by the Greek letters α, β, γ, δ, δ', and ϵ, respectively, have been reported from room temperature to the melting point, which is only 640°C (1185°F). The stability ranges and some of the properties of the six solid phases are given in Table 8.6. The alpha phase, stable at ordinary temperatures, was at one time thought to be isomorphous with alpha-uranium, but this is not the case. The delta and delta prime phases of plutonium are remarkable in having negative temperature coefficients of expansion, a property which is apparently unique among metals.

8.59. The electrical resistivity, which is 145×10^{-6} ohm-cm for the alpha phase and about 105 to 110×10^{-6} ohm-cm for the other five solid phases, is higher than that for any other metallic element. This suggests that the thermal conductivity of plutonium metal is probably relatively low. Another remarkable fact about plutonium is that for no phase do both the coefficient of thermal expansion and the temperature coefficient of resistivity behave in a conventional manner. Thus for the alpha, beta, gamma, and epsilon phases, the former is positive, which is normal, but the latter is negative. On the other hand, for the delta and delta prime phases the electrical resistance increases with temperature, as is usual, but now the coefficient of thermal expansion is negative.

SEPARATION OF URANIUM ISOTOPES

General Principles of Gaseous Diffusion

8.60. Enriched uranium, i.e., uranium containing more than the natural amount (0.71 per cent) of uranium-235, is being used as fuel in a number of reactors and will probably be employed in many reactors of the future. It is consequently appropriate to give a short account here of the method used for performing the isotopic separation necessary for producing enriched uranium. Several procedures for concentrating the uranium-235 in natural uranium were tested, but all others were soon abandoned in favor of the so-called *gaseous-diffusion process*. This process has been in successful operation at Oak Ridge, Tennessee, since 1945 and is being adopted for the newer separation plants at Paducah, Kentucky, and Portsmouth, Ohio.*

8.61. The gaseous-diffusion method for the separation of isotopes makes use of the different rates at which gases (or vapors) of different molecular weight diffuse through a porous barrier consisting of a number of fine holes or pores.†

* For descriptions of the original (K-25) plant at Oak Ridge, see J. F. Hogerton, *Chem. Eng.*, **52**, No. 12, 98 (1945); P. C. Keith, *ibid.*, **53**, No. 2, 112 (1946).

† The passage of gas through fine pores, small in comparison with the mean free path of the molecules, is properly called *effusion* rather than diffusion. The description "gaseous diffusion" for the isotope separation process is thus not strictly correct.

For the separation of the isotopes of uranium the barriers contain hundreds of millions of pores per square inch, the average diameter of the pores being about two millionths of an inch. A lighter isotopic molecule will diffuse (or effuse) through such barriers faster than a heavier one so that a partial separation of the isotopes is possible. The gas which first passes through the barrier will be relatively richer in the lighter isotope, whereas that remaining will contain a correspondingly higher proportion of the heavier isotopic form.

8.62. For the separation of the isotopes of uranium by diffusion, the gaseous compound employed is the hexafluoride, UF_6. As stated in § 8.27, this substance is a solid at ordinary temperatures, but it has a high vapor pressure; thus, at 56.5°C (133.7°F), the normal sublimation temperature, the vapor pressure is equal to 1 atm, i.e., 760 mm Hg. From the standpoint of gaseous diffusion, the use of uranium hexafluoride has two main advantages. First, the atomic weight of fluorine (19.0) is relatively low, so that the molecular weight of the diffusing gas, which is inevitably high because of the atomic weight of uranium, will not be too high. As will be evident shortly, the lower the molecular weight, the better the separation efficiency under given conditions. Second, since fluorine consists of a single species, the only isotopes separated in the diffusion process are those of uranium. The main disadvantages of uranium hexafluoride are its marked corrosive action on many metals and its ready interaction with moisture to form solid uranyl fluoride (UO_2F_2).

8.63. The extent of separation of isotopes in any process can be expressed by means of the *separation factor;* in the present case this is defined as the ratio of uranium-235 to uranium-238 atoms in the enriched state to that in the residual state. The rate of diffusion (or effusion) of a gas is inversely proportional to the square root of its molecular weight, and hence it can be shown that the theoretical separation factor, α^*, is given by

$$\alpha^* = \sqrt{\frac{M(\text{heavy})}{M(\text{light})}}, \tag{8.63.1}$$

where M(heavy) and M(light) are the molecular weights of the heavier and lighter isotopic diffusion species, respectively. Natural uranium consists essentially of uranium-238 and uranium-235; there is a small proportion of uranium-234 (§ 1.47), but this may be ignored for the present purpose. Hence, for uranium hexafluoride as the diffusing gas, M(heavy) is $(238) + (6 \times 19) = 352$, whereas M(light) is $(235) + (6 \times 19) = 349$. The theoretical separation factor is consequently

$$\alpha^* = \sqrt{\frac{352}{349}} = 1.0043.$$

The theoretical *enrichment factor*, defined as $\alpha^* - 1$, is therefore 0.0043 for uranium-235 in uranium hexafluoride.

8.64. Since the separation factor is so close to unity, the degree of enrichment in any diffusion stage is very small, but the effect can be multiplied by making

use of a "cascade" consisting of a number of stages, a few of which are indicated in Fig. 8.6. The feed gas, i.e., ordinary uranium hexafluoride in this case, enters at about the middle of the cascade. The diffusion stages above the feed are often referred to as the "enriching section," and those below as the "stripping section."* These terms imply that the gas moving into the upper stages becomes enriched in uranium-235, whereas that moving into the lower stages is impoverished in (or stripped of) this isotope.

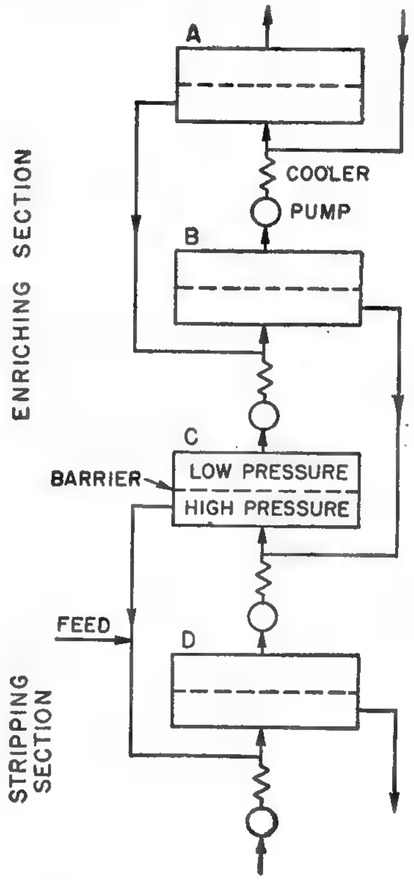


FIG. 8.6. Stages in diffusion cascade

8.65. Consider the stage marked B; the gas enters from the bottom and about half is allowed to pass through the porous barrier. The diffusate, which is richer in the lighter isotope, then flows up into stage A. Here it undergoes further diffusion, so that the diffusate contains a still higher proportion of this isotope. Turning attention once again to stage B, it is seen that the gas which has not diffused, and which is consequently somewhat impoverished in the lighter species, is pumped to stage C where it joins the diffusate from stage D. The gas then undergoes diffusion in stage C, the diffusate going on to stage B, while the residual gas returns to stage D to be recycled. In this way the gas moving upward through the cascade becomes increasingly richer in the lighter isotope, whereas that moving downward contains increasing proportions of the heavier isotope.

THE STAGE SEPARATION FACTOR

8.66. The theoretical treatment employed in the design of a cascade for gaseous diffusion is analogous to that used in connection with the separation of the more and less volatile constituents of a liquid mixture by fractional distillation.† Thus each stage of the cascade may be compared with a tray of a bubble-plate distilling column. In the diffusion cascade the gas is separated at each

* The terms "above" and "below," or "up" and "down," as used here and subsequently, are intended to have a purely figurative significance. The actual gas flow may be from right to left and vice versa. However, the use of "up" and "down" as directions permits a comparison to be made between a cascade and a fractionation column (§ 8.66).

† J. Schacter and G. A. Garrett, Report AECD-1940; G. A. Garrett, Report AECU-2537, p. 41; see also K. Cohen, "The Theory of Isotope Separation," National Nuclear Energy Series, Div. III, Vol. 1B, McGraw-Hill Book Co., Inc., 1951; *Nucleonics*, 2, No. 6, 3 (1948).

stage into an upflowing fraction enriched in the lighter isotope and a downflowing fraction depleted in this species. Similarly, in each stage of a fractionating column, a mixture is separated into an upflowing vapor fraction enriched in the more volatile constituent and a downflowing liquid fraction containing relatively more of the less volatile substance. Just as each diffusion stage is fed both by the upflow of enriched gas from the next lower stage and by the downflow of impoverished gas from the stage above, so each plate in a distilling column is fed by vapor from below and liquid from above.

8.67. The details of the calculations used in the development of an efficient diffusion cascade are too complicated to be given here. However, some relatively simple concepts will be presented so as to provide an indication of the order of magnitude involved. For any stage, e.g., the nth, of a diffusion cascade, the stage separation factor, which is equivalent to the relative volatility in fractional distillation, is given, in accordance with the definition in § 8.63, by

$$\alpha_n = \frac{y_n/(1-y_n)}{x_n/(1-x_n)} = \frac{y_n(1-x_n)}{x_n(1-y_n)}, \tag{8.67.1}$$

where y_n is the (atomic) fraction of the lighter isotope in the (enriched) upflow and x_n is the (atomic) fraction in the (depleted) downflow at the nth stage (Fig. 8.7). Thus, $y_n/(1-y_n)$ is the ratio of lighter to heavier isotope in the upflowing stream, whereas $x_n/(1-x_n)$ is that in the downflowing stream at the nth diffusion stage.

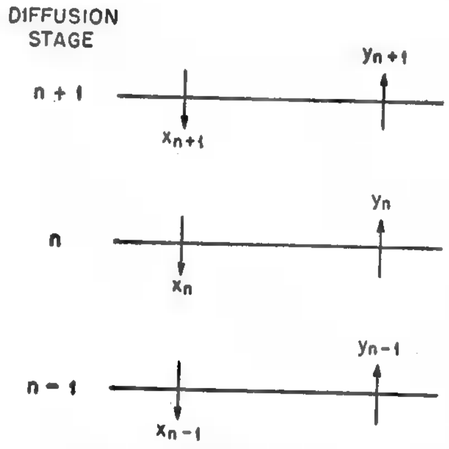


FIG. 8.7. Fraction of lighter isotope in enriched (upflowing) and depleted (downflowing) streams

8.68. The stage separation factor α_n, as defined by equation (8.67.1), differs from the theoretical separation factor α^*, as given by equation (8.63.1), for two reasons, in particular. As the gas stream on the high-pressure (under) side of the barrier flows from the entrance to the exit of a stage, there is a continuous depletion of the lighter isotope. The result is that x_n, the fraction of the lighter isotope in the downflow at the nth stage, is less than the average value along the barrier at this stage. The actual enrichment factor is consequently larger than the theoretical value to an extent depending on the "cut," i.e., the fraction of the gas entering the stage which actually passes through the barrier. If the cut is represented by θ, then it can be shown that

$$\alpha_n - 1 = \left(\frac{1}{\theta}\ln\frac{1}{1-\theta}\right)(\alpha^* - 1).$$

For practical purposes, θ may be taken as 0.5, i.e., half the gas entering the stage passes through the barrier, and so the cut correction, i.e., the factor in the first parentheses, is 2 ln 2, i.e., 1.38.

8.69. Although the cut effect tends to increase the enrichment factor above the theoretical value, other circumstances tend to reduce it. The most important is the fact that the gas pressure on the low-pressure side of the barrier is greater than zero; this introduces a resistance to diffusion which decreases the separation factor. In addition, as a result of the preferential diffusion of the lighter molecules, the concentration of this species close to the barrier on the high-pressure side is less than it is farther away.

8.70. Since the influence on the separation (or enrichment) factor of the two phenomena just considered is opposite to that of the cut effect, it will be assumed for the present treatment that the various effects cancel one another. For purposes of illustration, therefore, the stage separation factor will be taken to be equal to the theoretical value, i.e., 1.0043 for the separation of the isotopes of uranium by gaseous diffusion of the hexafluoride.

Theoretical Treatment of Gaseous-Diffusion Cascade

8.71. If L_n is the total amount of gas flowing upward through the barrier at the nth stage in a given time, generally referred to as the interstage flow rate, and P is the withdrawal rate of the product at the top of the cascade, then the total interstage downflow rate is $L_n - P$. Material balance considerations for the nth stage in the enriching section of the cascade then require that, in the steady state,

$$L_n y_n - (L_n - P)x_{n+1} = Px_p, \tag{8.71.1}$$

where x_p is the fraction of the lighter isotope in the product withdrawn at the top of the cascade. It can be seen, with the aid of Fig. 8.8, that the left side of this equation represents the net upward transport of the lighter isotope, and this must be equal to the rate at which this isotope is withdrawn, as indicated by the right side.

8.72. According to equation (8.67.1), the stage enrichment factor, $\alpha_n - 1$, is given by

$$\alpha_n - 1 = \frac{y_n - x_n}{x_n(1 - y_n)} \approx \frac{y_n - x_n}{x_n(1 - x_n)}, \tag{8.72.1}$$

the approximation of replacing $1 - y_n$ by $1 - x_n$ being justified by the fact that for the isotopes of uranium the extent of separation in any one stage is so small that $1 - x_n$ and $1 - y_n$ are not very different. Upon combining equations (8.71.1) and (8.72.1), it is found that

$$x_{n+1} - x_n = \frac{L_n}{L_n - P}\left[(\alpha_n - 1)x_n(1 - x_n) - \frac{P}{L_n}(x_p - x_n)\right]. \tag{8.72.2}$$

8.73. Because of the small change in the isotopic composition from one stage to the next, the stage number, n, and the concentration, x, may be treated as

continuous variables; the difference equation (8.72.2) may thus be replaced by a differential equation. Further, as will be evident shortly, the withdrawal rate P of the product is small in comparison with the interstage flow rate L, except at the top of the cascade, so that $L/(L - P)$ is essentially equal to unity. Upon making these two approximations, equation (8.72.2) becomes

$$\frac{dx}{dn} = (\alpha - 1)x(1 - x) - \frac{P}{L}(x_p - x), \tag{8.73.1}$$

for any stage in the enriching section of the cascade.

8.74. A similar equation can be derived for the variation of the isotopic concentration with the stage number in the stripping section. If W is the withdrawal rate of the waste at the bottom of the cascade, it is found that

$$\frac{dx}{dn} = (\alpha - 1)x(1 - x) - \frac{W}{L}(x - x_w), \tag{8.74.1}$$

where x_w is the fraction of the lighter isotope in the waste drawn off from the bottom of the stripping section (see Fig. 8.8).

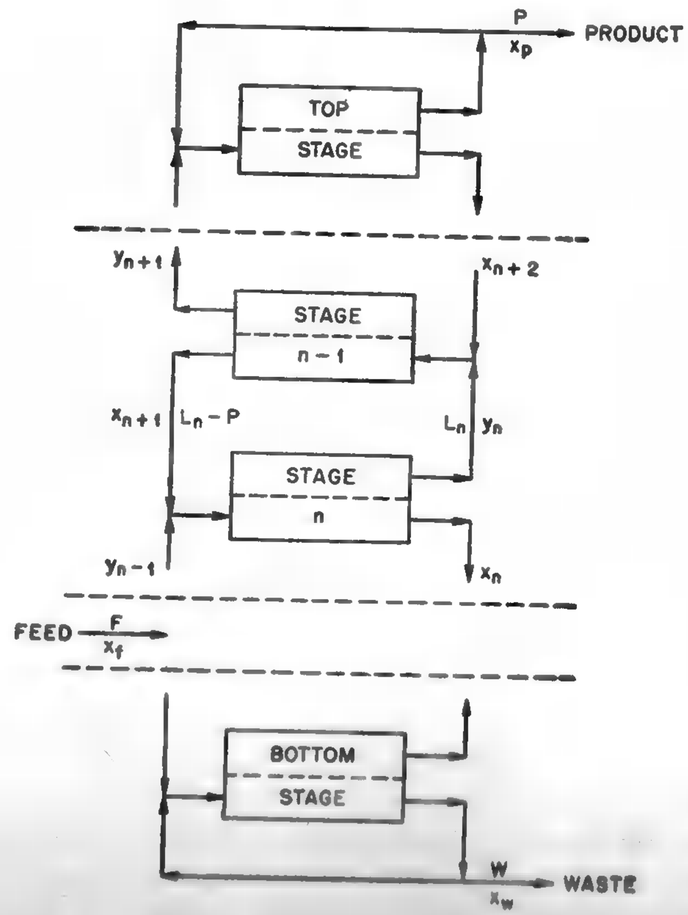


Fig. 8.8. Flow rates and compositions at various stages of cascade

8.75. The application of the differential equations derived above will be illustrated by reference to two special cases. These are (1) total reflux and (2) the ideal cascade. Before discussing these special applications, however, consideration will be given to the general relationships between the product and waste withdrawal rate, i.e., P and W, respectively, and the feed rate, F, at which the process gas, e.g., uranium hexafluoride, enters the feed stage of the cascade. The over-all material balance of the cascade requires that

$$F = P + W,$$

since the rate at which the feed gas enters the system must be equal to the sum of the rates of product and waste withdrawal in the steady state. A similar balance will apply to the quantities of the lighter isotope entering and leaving the cascade; this leads to

$$Fx_f = Px_p + Wx_w,$$

where x_f is the fraction of the lighter isotope in the feed gas, and x_p and x_w are, as before, the corresponding fractions in the product and waste, respectively. From these two equations the magnitude of the feed and waste rates, relative to the rate of product withdrawal, can be determined for any specified conditions.

Example Calculate the feed and waste rates for a diffusion cascade, fed with natural uranium hexafluoride, in which the (atomic) fraction of uranium-235 in the product is 0.90 and that in the waste is 0.002.

For the natural uranium feed, x_f is 0.0071, and since it is postulated that x_p is 0.90, and x_w is 0.002, it follows that

$$0.0071F = 0.090P + 0.002W.$$

Further, since $W = F - P$,

$$0.0071F = 0.090P + 0.002(F - P),$$

so that

$$F = 176P.$$

The feed rate is thus 176 times the rate of product withdrawal. The rate of waste removal, at the bottom of the stripping section of the cascade, is 175 times the rate at which the product is withdrawn at the top of the enriching section.

Total Reflux

8.76. A cascade is said to operate under conditions of total reflux when there is no product withdrawal, i.e., when P is zero. In these circumstances equation (8.73.1) becomes

$$\frac{dx}{dn} = (\alpha - 1)x(1 - x). \tag{8.76.1}$$

If this is integrated from x_f, the concentration of the lighter isotope in the feed, to any arbitrary concentration x, then the number of the stage in the enrichment section at which this concentration is attained is seen to be given by

$$n_{\min} = \frac{1}{\alpha - 1} \ln \frac{x(1 - x_f)}{x_f(1 - x)}, \tag{8.76.2}$$

the stage separation factor being assumed to be constant and independent of the stage number. The number of stages required to produce a specified enrichment is less at total reflux than under any other conditions. Consequently the subscript "min" has been added, to indicate that this is a minimum value for the number of stages.

8.77. It may be noted that equation (8.76.2) is also applicable to the stripping section of a cascade with total reflux, i.e., with W equal to zero. This equation is quite general, provided the feed point is designated as the stage for which $n = 0$; stages in the enriching section then have positive values of n and those in the stripping section have negative n values.

8.78. The minimum number of stages required to enrich the gas from the feed concentration x_f to the desired concentration x_p in the product is obtained by substituting x_p for x in equation (8.76.2). Similarly, the minimum number of stages in the stripping section is given by substituting x_w, the prescribed concentration in the waste stream, for x in this equation.

Example Determine the minimum number of enriching, stripping, and total stages required to convert a feed gas of natural uranium hexafluoride into a product in which the fraction of the lighter isotope is 0.90 and a waste stream in which the fraction is 0.002.

Since the feed material is natural uranium, x_f is 0.0071, and the specified values of x_p and x_w are 0.90 and 0.002, respectively. The stage enrichment factor will be taken as the theoretical value, i.e., 1.0043. Hence, from equation (8.76.2),

$$\begin{aligned}\text{Min. no. of enrichment stages} &= \frac{1}{\alpha - 1} \ln \frac{x_p(1 - x_f)}{x_f(1 - x_p)} \\ &= \frac{1}{0.0043} \ln \frac{(0.90)(0.9929)}{(0.0071)(0.1)} \\ &= 1660\end{aligned}$$

and

$$\begin{aligned}\text{Min. no. of stripping stages} &= \frac{1}{\alpha - 1} \ln \frac{x_w(1 - x_f)}{x_f(1 - x_w)} \\ &= \frac{1}{0.0043} \ln \frac{(0.002)(0.9929)}{(0.0071)(0.998)} \\ &= -295.\end{aligned}$$

The minimum numbers of stages are thus 1660 for the enriching section and 295 for the stripping section, making a total of 1955 stages.

8.79. It can be readily seen from the foregoing arguments that the minimum total number of stages is given by the expression

$$N_{\min} = \frac{1}{\alpha - 1} \ln \frac{x_p(1 - x_w)}{x_w(1 - x_p)}. \tag{8.79.1}$$

From this equation calculations have been made of the minimum number of stages required to yield product streams (x_p) containing 0.50, 0.90, 0.95, and 0.99, respectively, of uranium-235 for three different values 0.005, 0.002, and 0.001, respectively, of x_w in the waste stream. The results are recorded in Table 8.7. The percentage recovery of uranium-235 is equal to $100(Px_p/Fx_f)$ and this is given for each of the three values of x_w; the ratio P/F (or F/P) is readily determined from the material balance equations in § 8.75.

TABLE 8.7. CALCULATED MINIMUM NUMBERS OF STAGES IN DIFFUSION CASCADE FOR URANIUM ISOTOPE SEPARATION

x_p	$x_w = 0.005$	$x_w = 0.002$	$x_w = 0.001$
0.50	1240	1450	1610
0.90	1750	1950	2120
0.95	1920	2130	2290
0.99	2300	2520	2670
Recovery, %	30	72	86

8.80. From the data in the table it is evident that the production of material highly enriched in uranium-235 from natural uranium by gaseous diffusion of the hexafluoride involves a minimum of several thousand stages. The largest numbers of stages are, in general, required for the higher degrees of enrichment; thus approximately the same number of stages is needed to increase the fraction of uranium-235 in the product from 0.50 to 0.90 as is necessary to raise it from 0.90 to 0.99. In the design of a diffusion plant, the increased enrichment and percentage recovery resulting from an increase in the number of stages must be weighed against the cost of constructing and operating the plant.

The Ideal Cascade

8.81. While the foregoing calculations give some idea of the very large number of stages required to produce appreciable enrichment of uranium-235, they refer to a hypothetical (total reflux) system from which there is essentially no withdrawal of product or waste. A case of more practical interest which is susceptible of relatively simple treatment is that of the so-called "ideal" cascade. Such a cascade is defined as one in which the (enriched) gas stream entering any stage from below has the same composition as the (depleted) gas stream entering it from above. In general, for the $(n + 1)$th stage, the condition for an ideal cascade is seen from Fig. 8.8 to be

$$y_n = x_{n+2}.$$

The special interest of the ideal cascade lies in the fact that, for a given value of the stage separation factor, such a cascade has the minimum barrier area and the minimum pumping power consumption for specified feed and product concentrations.

8.82. By substituting x_{n+2} for y_n in equation (8.67.1), it follows that

$$\frac{x_{n+2}}{1 - x_{n+2}} = \alpha \frac{x_n}{1 - x_n},$$

the value of the stage separation factor being assumed independent of the stage number. If the enrichment between the stages n and $n + 1$ is taken to be the same as that between $n + 1$ and $n + 2$, then

$$\frac{x_{n+1}}{1 - x_{n+1}} = \sqrt{\alpha} \frac{x_n}{1 - x_n}$$

$$\approx [1 + \tfrac{1}{2}(\alpha - 1)] \frac{x_n}{1 - x_n}. \tag{8.82.1}$$

8.83. Since x_n and x_{n+1} are not greatly different, because of the low enrichment per stage, it can be shown from equation (8.82.1) that, to a good approximation,

$$x_{n+1} - x_n = \tfrac{1}{2}(\alpha - 1)x_n(1 - x_n). \tag{8.83.1}$$

Finally, assuming, as before, that x and n are continuously variable, this can be written as

$$\frac{dx}{dn} = \tfrac{1}{2}(\alpha - 1)x(1 - x). \tag{8.83.2}$$

Upon comparing this expression with equation (8.76.1) it is seen that the enrichment (or stripping) rate per stage in the ideal cascade is just half that for the total reflux condition. Consequently the number of stages required to achieve a specified concentration, x, is twice the minimum value given by equation (8.76.2); thus,

$$n_{\text{ideal}} = 2n_{\min} = \frac{2}{\alpha - 1} \ln \frac{x(1 - x_f)}{x_f(1 - x)}.$$

Similarly, the total number of stages in enriching and stripping sections is expressed by

$$N_{\text{idcal}} = 2N_{\min} = \frac{2}{\alpha - 1} \ln \frac{x_p(1 - x_w)}{x_w(1 - x_p)}.$$

For identical conditions of product and waste concentrations, the number of stages in the ideal cascade is twice the corresponding (minimum) value in Table 8.7.

8.84. Upon combining equation (8.83.2) for the ideal cascade with the general equation (8.73.1), it is seen that

$$\tfrac{1}{2}(\alpha - 1)x(1 - x) = \frac{P}{L}(x_p - x),$$

and hence

$$L_{\text{ideal}} = \frac{2P}{\alpha - 1} \cdot \frac{x_p - x}{x(1 - x)}. \tag{8.84.1}$$

From this expression can be calculated the interstage flow rate which must be

maintained in an ideal cascade at any particular stage of the enriching section, where x is the concentration of the lighter isotope in the downflowing stream. The corresponding equation for the interstage flow rate in the stripping section is

$$L_{\text{ideal}} = \frac{2W}{\alpha - 1} \cdot \frac{x - x_w}{x(1 - x)}. \tag{8.84.2}$$

It will be noted from these equations that the interstage flow rate in an ideal cascade varies from one stage to another. It is evidently greatest at the stage where the feed enters and then decreases steadily both in the enriching and stripping sections as the product and waste ends, respectively, are approached.

Example Determine the interstage flow rate at the feed stage in an ideal gaseous diffusion cascade, using natural uranium hexafluoride as feed, for a product having a uranium-235 fraction of 0.90.

Taking α as 1.0043, x_p as 0.90, and x, which in this case is x_f, as 0.0071, it follows from equation (8.84.1) that

$$L_{\text{ideal}} = \frac{2P}{0.0043} \cdot \frac{0.90 - 0.0071}{(0.0071)(0.9929)}$$
$$= 58{,}900P.$$

The flow rate at the feed stage is thus nearly 59,000 times the rate of product withdrawal.

8.85. It is important to understand that the interstage flow rate calculated in the foregoing example does not represent the rate at which the feed gas enters the cascade. This, as may be seen from the example in § 8.75, is much less than the flow rate at the feed stage. The latter represents the total amount of gas which must flow through the barrier at the feed stage in order to achieve the desired enrichment. From the engineering point of view its significance lies in the fact that it provides a measure of the total pumping power which must be expended at this stage. The same is true, of course, for all the stages in the cascade.

8.86. In order to indicate how the interstage flow rate (per unit product rate) and the concentration of uranium-235 vary with the stage number, calculations have been made, using equations (8.84.1) and (8.83.1), respectively, for an ideal cascade in which diffusion of gaseous uranium hexafluoride occurs. The composition of the product is 0.80 of uranium-235, and the waste contains the fraction 0.00355 of this isotope, which is half the amount present in the feed gas. The results are shown graphically in Fig. 8.9.* The number of stages in the enrichment section is 2940 and that in the stripping section is 320, making a total of 3260 stages. The feed rate is 224 times the rate of product withdrawal, and the recovery is just over 50 per cent.

8.87. The area under the interstage flow-rate curve gives the total flow rate for all the stages in the ideal cascade and is, consequently, a measure of the total pumping power for the whole cascade. An analytical expression for this total

* G. A. Garrett, *loc. cit.*

flow rate can be derived by using equations (8.83.2) and (8.84.1) to write an equation for $L\,dn$, and then integrating over all stages from waste to product. The result is

$$\text{Total interstage flow rate} = \frac{4}{(\alpha - 1)^2}[PV(x_p) + WV(x_w) - FV(x_f)], \tag{8.87.1}$$

where $V(x)$ is a function defined by

$$V(x) \equiv (2x - 1) \ln \frac{x}{1 - x}.$$

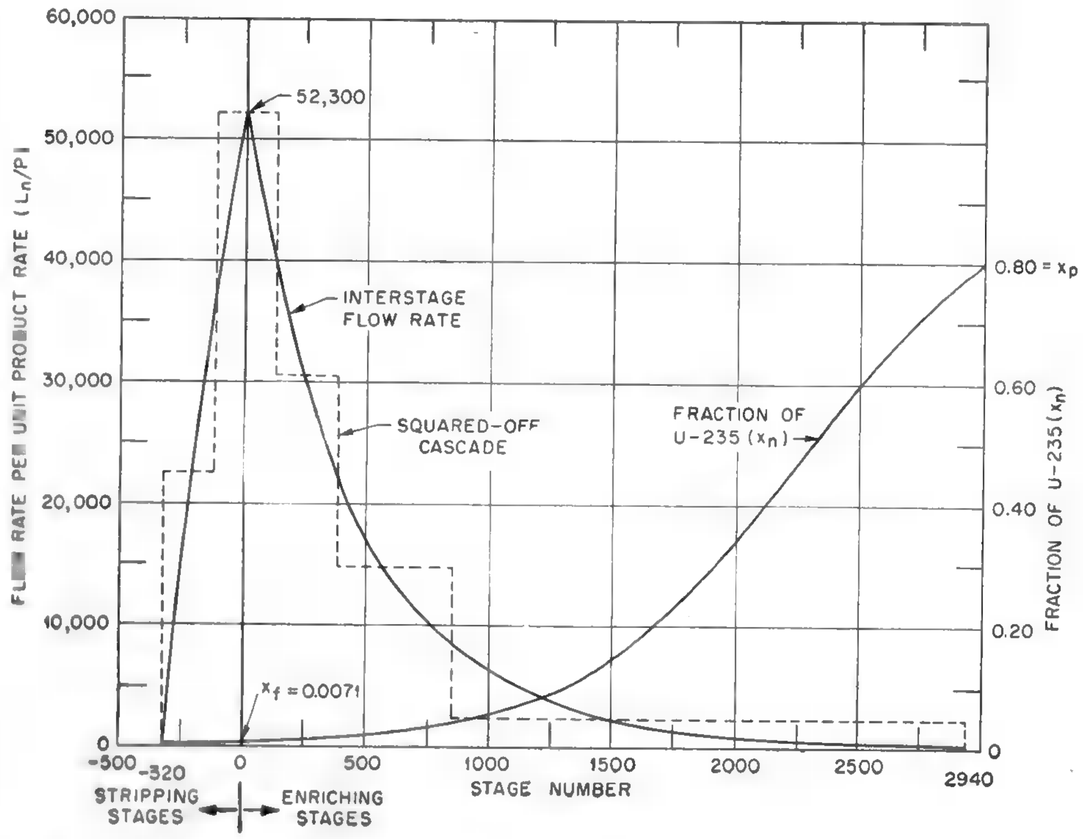


FIG. 8.9. Variation of interstage flow rate and fraction of uranium-235 with stage number (G. A. Garrett)

In the case to which Fig. 8.9 applies, the total interstage flow rate is found to be $34.4 \times 10^6 P$. In the design of a gaseous diffusion plant it is desirable to minimize the total flow rate in order to decrease the power consumption. This may be done by adjustment of the variable parameters in equation (8.87.1) within the limitations set by production specifications.

Squared-off Cascade

8.88. In spite of the virtues of requiring a minimum barrier area and minimum pumping power, the ideal cascade is not a practical one, since the flow rate and dimensions of every stage are different from those of every other stage. Obviously, a design involving several thousand different stages is not a practical possibility. The difficulty is overcome by using the device of what is called a "squared-off" cascade, as indicated by the broken lines in Fig. 8.9. It is seen that this is, in a sense, an approximation to the ideal cascade.

8.89. The squared-off cascade consists of a limited number of square cascades, in each of which all stages are the same size and have the same interstage flow rate. In the case depicted in Fig. 8.9, for example, there are four square cascades in the enriching section and two in the stripping section. The stages, i.e., vessels and associated pumping equipment, are largest in the square cascade where the feed enters, since the interstage flow rate is highest here. There are then successive decreases in size as the product and waste ends are approached.

8.90. The total number of stages in a squared-off cascade may be less than in an ideal cascade for the same product and waste concentrations. However, the total power requirement and the barrier area are larger in the squared-off cascade. Although the latter would thus be less economical in these respects, it is, of course, decidedly less expensive to construct than an ideal cascade in which every stage is different.

Reactor Fuel Cycles

8.91. In concluding this section dealing with uranium-235, and with reactor fuels in general, it is of interest to see how the various materials fit into the overall pattern of reactor design (see Fig. 8.10). From the long-range standpoint, two general phases may be expected in the utilization of fissionable and fertile species in reactors for power purposes. It should be understood that existing reactors do not necessarily fit into the scheme outlined below. First, because some are designed to produce plutonium for weapons, without regard to the energy released. And second, because others are of an experimental character, whose purpose is to provide information to be utilized in the design of practical systems for nuclear power production.

8.92. In the first phase, when relatively pure fissionable materials are in short supply, natural (or slightly enriched) uranium will be used as the reactor fuel. The spent fuel elements will be processed to recover the plutonium-239 present and the residual depleted uranium. The latter can then either be brought up to its former state of enrichment by the addition of uranium-235 (not shown in the figure) or it can be utilized as the fertile component of a breeder-type reactor using highly enriched or essentially pure fissionable material as fuel. These reactors represent the second phase of development. Originally they may be high-efficiency converters, utilizing uranium-235 as fuel and converting fertile uranium-238 and thorium-232 into fissionable plutonium-239 and uranium-233,

respectively. But ultimately these reactors of the second phase may be expected to be true breeders, in which the same fissionable species is formed as is consumed as fuel. In the early stages the breeding aspect, i.e., the production of more fissionable material than is used up, will be very important, but later, when the proportion of available fissionable to fertile materials has been greatly increased, this will become of lesser significance.

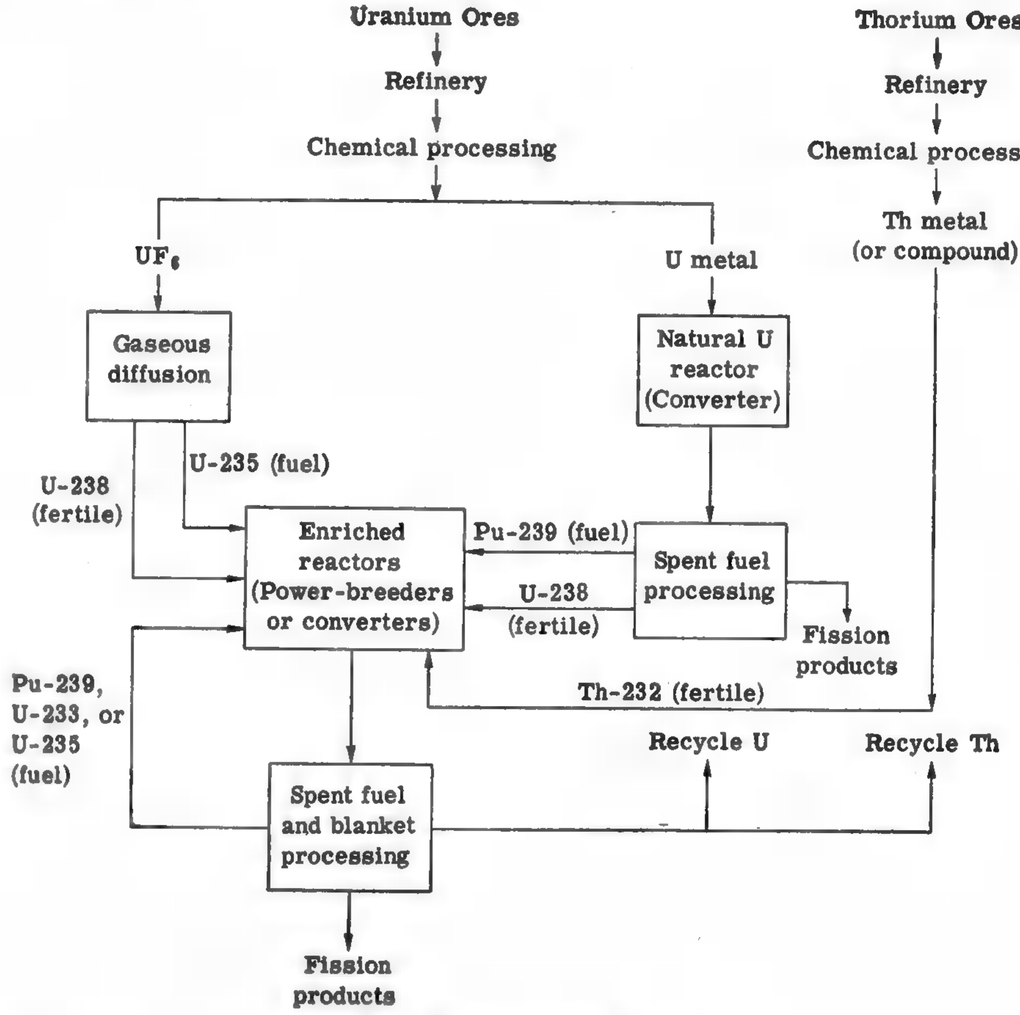


Fig. 8.10. Reactor fuel cycles

8.93. Because of the limitations set upon the extent of fuel consumption by burn-up, fission product poisons, and so on, recycling of the fuel elements through processing plants will be necessary from time to time. The role of such plants and the application of the materials obtained from them is shown in Fig. 8.10. These plants, as well as the gaseous-diffusion plant for the separation of the isotopes of uranium, are seen to be essential parts of the reactor fuel cycles.*

* J. F. Flagg and E. L. Zebroski, *Scientific American*, **187**, No. 1, 62 (1952); Report AECU-1062.

RADIATION EFFECTS IN SOLIDS*

General Effects of Nuclear Radiations

8.94. The radiations in a nuclear reactor consist of alpha and beta particles, gamma radiations, neutrons, fission fragments, and, possibly, protons. Although the fission fragments are not strictly nuclear radiations, they are not essentially different from alpha particles and protons, both of which are charged atomic particles and, except in degree, their effects are the same.

8.95. It was noted in Chapter II that nuclear radiations are able to cause atoms and molecules to become ionized or electronically excited, either directly or indirectly. These effects may lead to chemical changes, especially if the radiations interact with compounds containing covalent bonds, such as organic materials, water, and certain salts, e.g., nitrates. This aspect of the action of radiations will be considered later; for the present the discussion will be restricted to the physical changes produced in metals or in electronic conductors generally.

8.96. As far as the ionization and electronic excitation they produce are concerned, nuclear radiations have almost no permanent effect upon metals. The reason is that in a metal the electrons in the conduction band are able to accept very small amounts (quanta) of excitation energy, and so a considerable proportion of the energy of the radiation is lost in causing electronic excitation. Because of the large number of empty energy states available, the excited electrons rapidly lose their excess energy; this is taken up by the atoms of the metal and appears in the form of heat, i.e., as vibrational energy of the nuclei.

8.97. Unless they are extremely energetic, beta particles and gamma-ray photons produce little, if any, radiation damage in electronic conductors, such as metals and graphite, but this is not necessarily true for heavy nuclear particles, such as protons, neutrons, alpha particles, and fission fragments. As a result of elastic collisions, these particles may transfer appreciable amounts of energy to the nuclei of a solid. If the amount of energy transferred is sufficient to cause the nuclei to be displaced from their normal (or equilibrium) positions in the space lattice, physical changes of a more or less permanent character will be observed in the metal. This effect of nuclear radiation is sometimes referred to as "radiation damage."

8.98. Among the changes which are generally produced in metals by radiation are increase in electrical resistance, in thermal resistance, in hardness, and tensile strength.† It appears that after exposure to radiations metals and alloys have higher yield strengths, lower percentage elongations, and somewhat higher ductile-brittle transition temperatures. On the whole, the effects are less marked

* For reviews, see J. C. Slater, *J. Appl. Phys.*, **22**, 237 (1951); G. J. Dienes, *Ann. Rev. Nuc. Sci.*, **2**, 197 (1953); *J. Appl. Phys.*, **24**, 666 (1953); D. S. Billington, Report AECU-2900, p. 120.

† D. S. Billington and S. Siegel, Report AECD-2810; *Metal Prog.*, **58**, 847 (1950); C. R. Sutton and D. O. Loosor, *Chem. Eng. Prog. Symposium Series*, No. 12, **50**, 208 (1954); D. O. Loosor, *Materials and Methods*, **40**, 110 (1954).

on hardened metals than in the annealed condition. The effects of radiation are generally smaller at elevated temperatures, and, as will be seen shortly, there are cases in which radiation produces no changes except at very low temperatures. Another aspect of temperature is that the effects produced by radiation can generally be removed ("annealed") by raising the temperature.

8.99. Conflicting results concerning the effect of radiation on the creep rate of metals have been reported, but this situation may have arisen from the failure to take into account the influence of temperature. It now appears that at low temperatures neutron bombardment causes the creep rate to decrease below the normal value, but above a certain critical temperature the creep rate may increase under the influence of radiation.

8.100. That internal changes in the crystal lattice result from the action of nuclear radiation has been proved by X-ray measurements on "ordered" samples of the gold-copper alloys $AuCu_3$ and AuCu. It is of interest to note in this connection that, whereas ordered specimens of $AuCu_3$ exhibit a considerable increase in electrical resistivity upon exposure to neutrons, disordered samples of this alloy show no such increase.

8.101. Attempts have been made to relate the behavior of metals under irradiation to more conventional phenomena, such as cold-working, solution- or alloy-hardening, and precipitation-hardening. Because the physical changes produced by cold-work are often similar to those resulting from exposure to radiation, it was thought that these two effects might be analogous. Some support for this view appeared to come from the fact that cold-worked metals exhibit little further change upon irradiation. However, various considerations, including differences in the X-ray patterns, indicate that the internal effects of cold-work and radiation are not similar. From a study of the critical shearing stress in single crystals of copper, it has been concluded that if an analogy does exist between changes caused by radiation and other phenomena it must be to solution- or alloy-hardening rather than to cold-working.

8.102. There is little doubt that the effect of nuclear radiations on metals results from localized distortion or damage to the crystal lattice. Two models have been proposed to account quantitatively for the action of radiation. These are (1) the atomic displacement theory and (2) the thermal-spike concept. Although these points of view are not mutually exclusive, it is convenient to treat them separately.

The Atomic Displacement Theory*

8.103. If, as the result of an elastic collision between the relatively heavy nuclear particle and an atomic nucleus, the energy transferred to the latter exceeds a certain minimum value (usually assumed to be about 25 ev for a metal), the struck ("knocked on") atom may be displaced from its normal or equilibrium

* F. Seitz, Irradiation Physics Lecture Notes, 1947 (unpublished); *Discussions Faraday Soc.*, **5**, 271 (1949).

position in the solid lattice.* If several atoms in close proximity are displaced in this manner, some will merely be transferred from one equilibrium position to another in the lattice. However, if a knocked-on atom is unable to find a vacant equilibrium site, its final location will be a nonequilibrium position. An atom of this kind is said to occupy an interstitial site and is known as an interstitial atom or, in brief, as an "interstitial." For each interstitial atom produced by the action of radiation, there must be a corresponding vacant site in the lattice, possibly at some distance away. The net result is thus a more or less permanent defect in the solid, which, if sufficiently common, may be accompanied by a change in its physical properties.†

8.104. In many cases the kinetic energy transferred to the knocked-on atom, generally referred to as a "knock-on," is so large that it can, in turn, produce secondary knock-ons by elastic collision with other atoms in the solid. The secondary knock-ons which have sufficient energy may, in turn, produce tertiary knock-ons, etc. Since the knock-ons are frequently ionized, due to what is called "impact ionization," they will lose some of their energy by causing ionization and electronic excitation. Consequently knock-ons beyond the third order can be neglected since the probability of transferring sufficient energy in an elastic collision to produce a further knock-on is then very small.

8.105. By making estimates of the rate of energy loss in the various processes in its passage through a solid, the total number of knock-ons produced by a 2-Mev neutron has been computed in a few cases. The results are quoted in Table 8.8. The number of knock-ons calculated for beryllium is seen to be

TABLE 8.8. CALCULATED NUMBER OF KNOCK-ONS FOR A 2-MEV NEUTRON

Material	*Number*
Beryllium	450
Beryllium oxide	2580
Graphite	1870
Aluminum	6030

considerably less than for aluminum; this is because the energy lost in causing electronic excitation is much larger in beryllium. The difference in behavior of beryllium and its oxide, as shown by the data in the table, is attributed to the differences in energy transfer in metals and nonmetals. It should be noted that the calculations give the total number of atoms displaced by a 2-Mev neutron. Not all of these will lead to lattice defects since many of the displaced atoms will undoubtedly move to vacant equilibrium positions.

* The displacement of atoms from their positions in the crystal lattice as a result of fast-neutron impact was predicted by E. P. Wigner in 1942. It has consequently been called the "Wigner effect."

† A lattice vacancy and an interstitial atom when considered as a unit are often called a "Frenkel defect."

The Thermal-Spike Concept*

8.106. A fission fragment will dissipate essentially all its energy and a fast neutron will lose a large proportion of its energy within a very small volume. This energy, no matter whether it is removed as atomic translation or vibration or as electronic excitation, will be rapidly converted into heat. The result will consequently be the formation of a small region of high temperature; this is the so-called "thermal spike." It has been estimated that the spike will include some 5000 to 10,000 atoms and that it attains a temperature of 700 to 1200°C (1290 to 2190°F) for a period of about 10^{-10} sec. At the high temperatures of the thermal spike, distortion of the lattice, equivalent to local fusion or even vaporization, is to be expected. In the very rapid cooling, resulting from the conduction of heat to the surroundings, a certain amount of the lattice distortion will be "frozen," since there will not be sufficient time for all the atoms to be relocated in equilibrium positions. In addition to the lattice defects produced in this manner, there will inevitably be microscopic changes due to diffusion.

8.107. Although both the atomic displacement theory and the thermal-spike concept have been used individually to account for the disordering effects of radiation on certain alloys in the ordered state, it is not impossible that they refer to two more or less distinct phenomena. Thus both might be operative simultaneously. For example, near the end of its path a nuclear particle might not have sufficient energy to displace an atom completely from its lattice position, but might, nevertheless, be capable of causing a localized region of high temperature. The operation of the displacement and thermal-spike mechanisms at the same time would make it difficult to account quantitatively for the observations, for one mechanism might interfere with or annihilate the effects of the other.

Radiation Damage Due to Impurity Atoms

8.108. Changes in the physical properties of metals due to the action of radiation may arise from a cause entirely different from those considered above, namely, the introduction of a new atomic species. When exposed to neutrons, for example, most elements undergo (n, γ) reactions. The immediate product of such a reaction is isotopic with the absorbing element, and so there should be no damage to the irradiated substance; but, if the product is radioactive, as is frequently the case, it will emit beta particles and be thereby converted into a different element. The result will be that an impurity atom is introduced into the original crystal lattice. If present in sufficient numbers, such impurity atoms will undoubtedly affect the physical properties of the material, thus leading to a type of radiation damage. It should be realized, however, that, whereas only one impurity atom can result from each neutron captured, a fast neutron may cause the displacement of a thousand or more nuclei as a result of energy

* F. Seitz, *loc. cit.*; S. Siegel, *Phys. Rev.*, 75, 1823 (1949); H. Brooks, unpublished.

transfer in elastic collisions. The changes due to (n, γ) reactions will thus be of a lower order than the radiation effects considered earlier.

Radiation Effects and Healing in Metals

8.109. It is generally accepted that, when an atom in a crystal lattice acquires a certain amount of energy, it is able to migrate from one site to another. The ease with which this migration occurs will presumably be greatest when the atom originally occupied a nonequilibrium (interstitial) site and moves into a vacant equilibrium site. Since it is probable that a metal which has been exposed to radiation contains both interstitial atoms and vacant sites, there should be a tendency for the former to move into the latter, i.e., for recombination to occur. In other words, as a result of migration in the lattice, there will always be some spontaneous healing of radiation damage of the defect type.

8.110. The rate of migration (or recombination) may be expected to increase with temperature, and hence so also should the rate of healing of radiation effects. The rate of healing (or annealing) may also be expected to increase with the total extent of the damage, since the larger the number of lattice defects the greater will be the probability that an interstitial atom will migrate into a vacant equilibrium site.

8.111. When a solid is irradiated, the net rate of production of damage in the lattice is equal to the rate of formation of lattice defects minus the rate of healing under the given conditions. Since the rate of healing will increase with the total damage, it is to be expected that, if a material is irradiated continuously at constant radiation density, a steady state will ultimately be attained when the rate of production of defects is exactly equal to the rate at which they are spontaneously healed by recombination. In this event the radiation effects are said to have become "saturated" or to have "reached saturation."

8.112. The total lattice damage at the saturation point may be expected to increase with the density (or flux) of the given radiation since this will increase the rate of formation of defects. Increase of temperature, on the other hand, will decrease the extent of the changes caused by radiation because of the increase in the rate of atomic migration and, hence, of healing. In agreement with anticipation, it has been found that the effects of nuclear radiation on metals is greater at low than at high temperatures. For example, radiation has very little effect on aluminum at ordinary temperatures, but changes have been observed when the metal is exposed at around −150°C (−240°F). Similarly, many of the changes produced in metallic copper by nuclear radiation at −170°C (−275°F) are reduced in magnitude when the material is maintained at room temperature.

8.113. The production of lattice defects and the consequent increase in the rate of atomic migration lead to some other interesting effects of radiation in metals. If a sample of $AuCu_3$ in the disordered state is maintained at 150°C (300°F) for several days there is no appreciable change in electrical resistivity.

However, when the material at the same temperature is exposed to neutrons in a reactor, a decrease of resistivity, indicating formation of the ordered state, is apparent in a few hours. At ordinary temperatures, the reverse change, i.e., from ordered to disordered state, occurs, as stated earlier. It has also been observed that the precipitation reaction in a copper-beryllium alloy commences at a lower temperature than normal under the action of neutrons. This is attributed to the enhancement of atomic migration in the lattice due to the effect of the radiation.

Radiation Effects in Nuclear Reactors

8.114. As far as the changes produced in reactor materials by radiations are concerned, the most important particles are fission fragments and neutrons. Although fission fragments have very high energies, their range is small and consequently such physical changes as they may cause will be confined almost entirely to the fuel element. These changes, in dimensions at least, may be very considerable, as is indicated by the photographs in Fig. 8.11; they show,

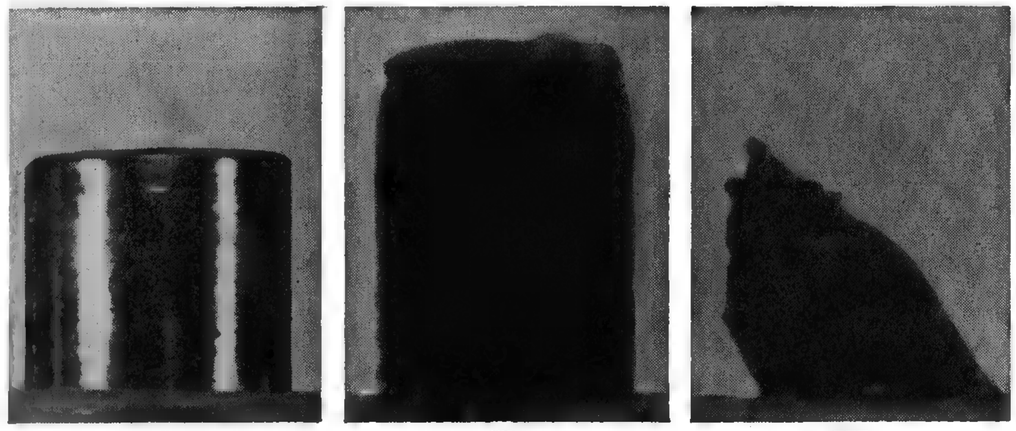


Fig. 8.11. Effect of reactor radiation on natural uranium: (I) before exposure, (II) limited exposure, (III) twice preceding exposure (Argonne National Laboratory)

first, an unexposed fuel element of uranium metal, and then two similar elements after increasing periods in a reactor. It will be immediately apparent that radiation damage can act as a very serious limitation upon the time of reactor operation. Considerable attention has been given to this matter, and it has been reported that the extent of the damage can be greatly decreased by suitable metallurgical treatment of the uranium.

8.115. It is generally accepted that the dimensional and other changes observed in irradiated fuel elements are largely due to fission fragments. Nevertheless, neutrons undoubtedly contribute to some extent to the over-all effects. Because of their lack of electrical charge, neutrons lose little, if any, of their

energy in producing ionization or electronic excitation. Thus essentially all the energy is lost in nuclear collisions, most of which, except for neutrons of high energy, are elastic in character (§ 2.103). Neutrons are thus capable of displacing many atoms in their passage through matter (cf. Table 8.8). The scattering mean free path of a neutron varies with its energy and with the scattering material, but in most substances the average distance traveled between successive elastic collisions is about 5 cm. The net (vector) slowing-down distance of a fission neutron in a poorly absorbing material is 10 to 20 cm. It would appear, therefore, that the radiation damage caused by neutrons will be less intense than that due to fission fragments and will be spread over a much larger volume.

8.116. Since fission fragments cannot get very far, owing to their short range, any changes that occur in the moderator must be attributed to the action of neutrons. In graphite, for example, it is known that exposure to neutrons affects the electrical resistance, the thermal conductivity, and the elasticity.* Such changes in the physical properties of the moderator can have a significant influence on reactor operation. One of the problems in reactor design is, therefore, to minimize the damage caused by radiation, to the moderator as well as to the fuel elements.

8.117. There is, of course, the possibility that neutrons may produce harmful effects in the structural material of the reactor. From the limited information at present available, it would appear that this is not a matter of great concern. However, the effects of extensive exposure to radiation, such as might occur in the continued operation of a power reactor, have not been definitely established. One of the main objectives in building the Materials Testing Reactor at Arco, Idaho, which normally operates at the high average flux of 2×10^{14} thermal neutrons/(cm^2)(sec), was to provide facilities for the study of radiation damage. As more information on this subject becomes available, it may be possible to choose the design and operating conditions of a reactor so as to minimize the effects of radiation.

Effects of Radiation on Nonmetallic Materials

8.118. In addition to graphite, which behaves in some respects like a metal, two groups of solid nonmetallic materials may be used in connection with a reactor; these are (1) plastics and (2) ceramics. The general effects of nuclear radiations on ceramics will be discussed in a later section dealing with these substances (§ 8.212), but the changes produced in plastic materials will be reviewed here.

8.119. Plastics, in the unfilled condition, are essentially pure organic compounds, and so it is to be expected that they will be badly affected by radiation, because of the breaking of the interatomic (covalent) bonds. This has been

* H. D. Smyth, "Atomic Energy for Military Purposes," U. S. Government Printing Office, 1945, § 7.24.

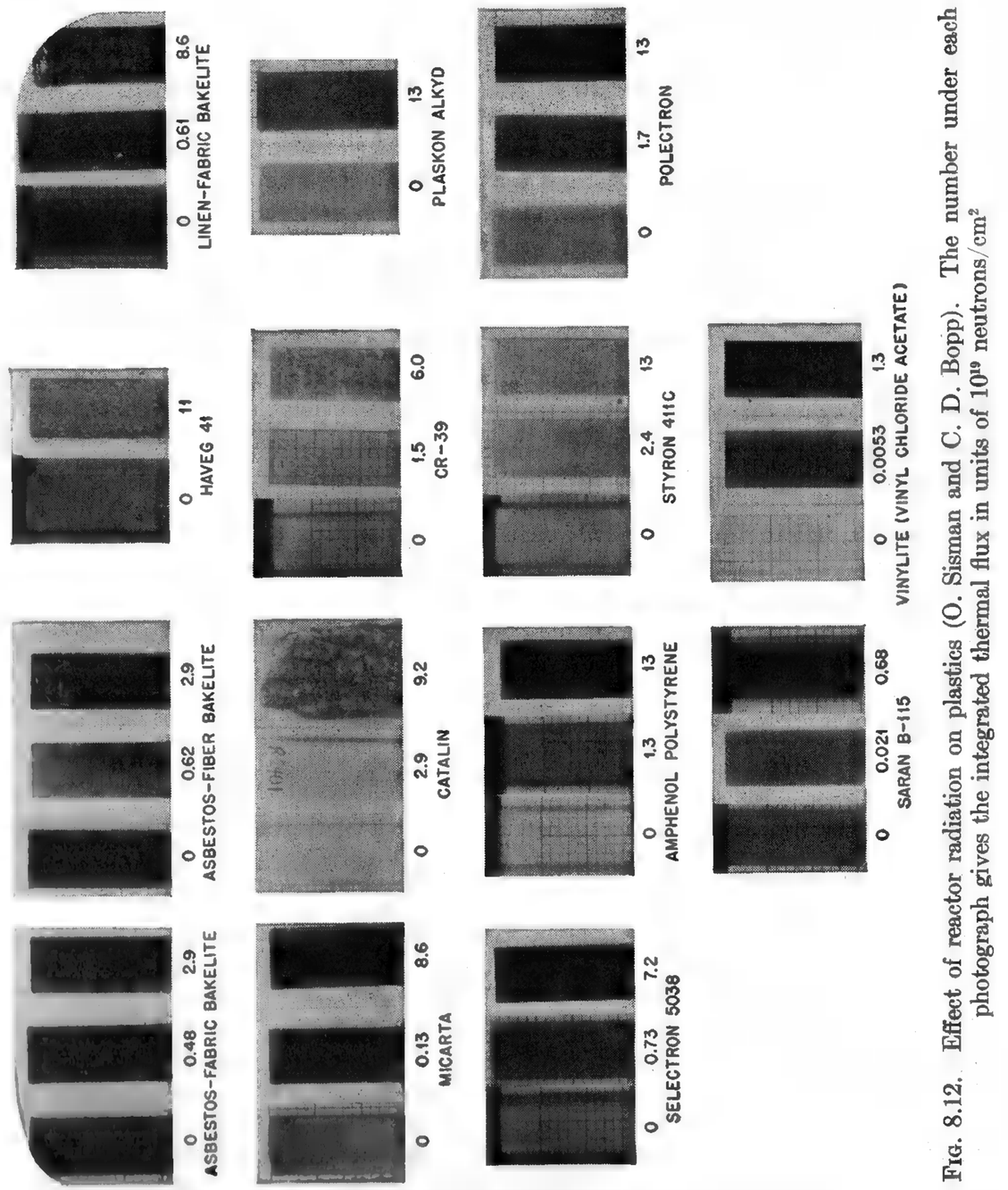


Fig. 8.12. Effect of reactor radiation on plastics (O. Sisman and C. D. Bopp). The number under each photograph gives the integrated thermal flux in units of 10^{19} neutrons/cm^2

found to be true in many cases, although mineral-filled plastics, e.g., those containing asbestos, are considerably more stable.

8.120. An extensive study has been made of the changes produced in a number of commercial plastics as the result of exposure in a nuclear reactor.* The radiation consisted mainly of thermal neutrons, although an appreciable proportion of neutrons of intermediate and high energies were also present. The gamma-radiation flux was about half that of the thermal neutrons. The effects of irradiation by an integrated thermal flux, i.e., the product of the thermal flux and the exposure time, of 10^{19} neutrons/cm^2 on six groups of the least-affected plastic materials are given in Table 8.9. The changes produced in various plastics are also shown by the photographs in Fig. 8.12. It is seen that certain mineral-filled plastics and unmodified polystyrenes are the most stable. Other plastic materials not mentioned in the table, e.g., phenolics (unfilled and cellulose filled), vinyl polymers, cellulose derivatives, and melamine and urea resins, were also investigated, but these proved, on the whole, to be affected to a greater extent.†

TABLE 8.9. EFFECTS OF INTEGRATED THERMAL NEUTRON FLUX OF 10^{19} NEUTRONS/CM^2 ON PLASTIC MATERIALS

Plastic Material	*Effect Observed*
Mineral-filled furan and mineral-filled phenolics	Little change except for darkening in color
Styrene polymers	Little change except for darkening in color
Modified styrene polymers	Impact strength and elongation decrease until the same as unmodified polystyrenes
Aniline formaldehyde and polyvinyl carbazole	Tensile strength decreases slightly
Nylon and polyethylene	Impact strength decreases but tensile strength increases; become very brittle
Mineral-filled polyester	Impact strength and tensile strength decrease about 50 per cent.

Radiation Effects on Nonmetallic Materials in Nuclear Reactors‡

8.121. Various nonmetallic materials, which are subject to the action of radiation, are used in some of the auxiliary components of a reactor. Mention may be made in particular of electrical insulators, capacitors, microswitches, and selsyn motors required for reactor instrumentation and control, and lubricants, hydraulic fluid, gaskets, and seals used in the heat-transfer system. Because of the instability of organic compounds, it is recommended that inorganic (ionic)

* O. Sisman and C. D. Bopp, Report ORNL-928; see also, D. S. Billington, Report AECU-2900, p. 120.

† It has been reported that in certain cases the physical properties of a plastic material have been improved upon exposure to radiation due to the formation of cross linkages between the polymer chains. The integrated flux employed in these instances was much less than would be involved in reactor operation.

‡ V. P. Calkins, *Chem. Eng. Prog. Symposium Series*, No. 12, 50, 28 (1954).

compounds be used wherever possible. Examples are mica and ionic ceramic capacitors, ceramic-covered resistors, and ceramic-bodied microswitches. Ionic ceramics or, possibly, mineral-filled plastics may be used for electrical insulation in selsyns and other motors, and for general electrical wiring. In view of the results recorded in Table 8.9, polystyrenes might be used as insulating materials, where the radiation flux is not too high, if their other physical properties are suitable.

8.122. In the heat-transfer system it would be desirable to fabricate gaskets, seals, couplings, etc., from metals whenever possible. In some cases ionic ceramics or other inorganic compounds could be used for certain components, e.g., for couplings. As lubricants, solids, such as graphite and molybdenum disulfide, might be applicable, since these are much more resistant to radiation than are the usual organic lubricants. Where a free-flowing oil is necessary, polyphenyl derivatives have been found to be appreciably more stable than the conventional hydrocarbons. In general, when the use of an organic compound is inevitable, care should be taken in the design of the component or the system to minimize the radiation exposure.

PRODUCTION AND PROPERTIES OF MODERATORS

General Review of Properties

8.123. It was seen in Chapter III that the only substances which might be used as moderators are ordinary water, heavy water (deuterium oxide), beryllium (either as metal, oxide, or carbide), and carbon (as graphite). The production and physical properties of these materials will be considered in turn. Since the characteristics of a good reflector for thermal neutrons are generally similar to those of a moderator, the descriptions given below may be regarded as applying to both moderators and reflectors. The important properties of five moderating materials are summarized in Table 8.10; beryllium carbide has not been included as its chemical reactivity with air and water seems to limit its usefulness. The densities of graphite and beryllium oxide vary with the method of production; the values given in the table are both less than the theoretical densities, but they may be taken to be typical of reactor materials. The abbreviations SDP and MR refer to slowing-down power and moderating ratio, respectively. In calculating these quantities the scattering cross sections used are those for epithermal neutrons, i.e., for neutrons having energies in excess of the thermal value, since they determine the rates at which the neutrons are slowed down. All other properties in the table refer to neutrons of thermal energy at ordinary temperatures.

Ordinary Water as Moderator

8.124. Ordinary water is attractive as a moderator because of its low cost, its excellent slowing-down power, and its small migration length for thermal neu-

TABLE 8.10. PROPERTIES OF MODERATORS AND THERMAL NEUTRON REFLECTORS

Properties	Ordinary Water	Heavy Water (99.75% D_2O)	Beryllium Metal	Beryllium Oxide	Graphite
Atomic (or molecular) wt.	18.0	20.0	9.01	25.0	12.0
Density, g/cm³	1.00	1.10	1.84	2.80	1.62
N (atoms or molecules/cm³)	3.3×10^{22}	3.3×10^{22}	1.2×10^{23}	6.7×10^{22}	8.1×10^{22}
σ_s barns (epithermal)	49	10.5	6.0	9.8	4.8
σ_a barns (thermal)	0.66	0.0026	0.009	0.0092	0.0045
Σ_s cm^{-1} (epithermal)	1.64	0.35	0.74	0.66	0.39
Σ_a cm^{-1} (thermal)	0.022	0.000085	0.0011	0.00062	0.00037
ξ	0.93	0.51	0.206	0.17	0.158
$\xi\Sigma_s$ cm^{-1} (SDP)	1.5	0.18	0.16	0.11	0.063
$\xi\Sigma_s/\Sigma_a$ (MR)	70	21,000	150	180	170
Diffusion coefficient, D cm	0.18	0.85	0.61	0.56	0.92
Diffusion length, L cm	2.88	100	23.6	30	50
Fermi age, τ cm²	33	120	98	110	350
Migration length, M cm	6.4	101	26	32	54

trons. On the other hand, its absorption cross section for neutron capture is relatively high, so that a critical system can be attained with water as moderator only if uranium enriched in the fissionable isotope is used as fuel. However, if enriched material is available, the small migration length of thermal neutrons in water makes it possible to design a reactor of relatively small size. The fact that water can be used as both moderator and coolant, as it is in several reactors (see Table 1.10), is an added advantage, but the water must be free from impurities since these not only capture neutrons but they may become radioactive as a result of (n, γ) reactions, thus constituting a hazard in the cooling system. Water of a high degree of purity is also desirable in order to minimize corrosion and scale formation. The decomposition of water by nuclear radiations, with the liberation of gaseous hydrogen and oxygen, is also often facilitated by the presence of ionic impurities (§ 8.142).

8.125. One of the main drawbacks to the use of water as a moderator is its relatively low boiling point. This means that, if it is to be utilized in high-temperature reactors, the pressures must be high, thus introducing fabrication and construction problems. For example, in the Pressurized Water Reactor (PWR), which is to be the first reactor designed to produce electrical power on a commercial basis, it is planned to operate at temperatures of 260 to 320°C (500 to 600°F) and pressures in the vicinity of 2000 psi.*

* "Report on the Five-Year Power Reactor Development Program Proposed by the Atomic Energy Commission," U. S. Government Printing Office, 1954, p. 10.

Heavy Water as Moderator*

8.126. Because of its satisfactory slowing-down power and exceptionally high moderating ratio (Table 8.10), heavy water is an excellent moderator and reflector. Under equivalent conditions, both the resonance escape probability and the thermal utilization are larger than when graphite is the moderator. Consequently, as seen in Chapter III, a heterogeneous natural-uranium reactor is smaller and requires considerably less fuel if heavy water is the moderator rather than graphite. In fact, a homogeneous system of natural uranium and heavy water can become critical, whereas such a mixture of natural uranium and graphite cannot (§ 3.139). Until recently, only limited quantities of heavy water were available, so that its use was restricted to laboratory reactors (see Table 1.10). With the development of improved production facilities, the situation is undergoing change. Heavy water is to be used as moderator in the production reactors at the Savannah River plant, and it will undoubtedly be available for other reactors in the future.

8.127. Pure deuterium oxide melts at 3.82°C (38.87°F) and boils at 101.42°C (214.56°F), the values, in each case, being a few degrees above the corresponding temperatures for ordinary water. High-temperature systems involving heavy water, like those using ordinary water, would therefore require high pressures. The density of heavy water is about 1.10 g/cm³ at room temperature.

Production of Heavy Water†

8.128. All natural waters contain about 1 part in 6500, i.e., about 0.015 per cent, of deuterium oxide, and heavy water is obtained by concentrating this very small proportion to any desired degree of purity. Several methods have been used to concentrate the deuterium in water; three in particular have shown prospects of successful application on a large scale. These are the electrolytic, distillation, and chemical-exchange methods. In addition, there is a possibility that an economical process may be developed for the separation of deuterium by the low-temperature distillation of liquid hydrogen.

8.129. The *electrolytic method* of producing heavy water depends upon the fact that, when an aqueous solution is electrolyzed, i.e., a current of electricity is passed through it, the hydrogen gas liberated at the cathode contains relatively more of the lighter than of the heavier isotope. The water remaining in the electrolytic cell is thus partially enriched in deuterium. For commercial purposes the electrolyte is usually an aqueous solution of sodium or potassium hydroxide or carbonate; the anodes are of nickel or nickel-plated steel, and the cathodes are made of nickel or steel. A steel electrolytic cell may serve as both

* W. H. McCorkle, *Nucleonics*, **11**, No. 5, 21 (1953).

† For reviews, see P. J. Selak and J. Finke, *Chem. Eng. Prog.*, **50**, 221 (1954); F. T. Barr, Report AECD-2871; "Production of Heavy Water," National Nuclear Energy Series, Div. III, Vol. 4F, McGraw-Hill Book Co., Inc., to be published, 1955.

container and cathode. By continued electrolysis it is possible to obtain heavy water approaching 100 per cent purity. Very large quantities of electrical energy are required to bring about this degree of concentration, and such a procedure, starting from ordinary water, is possible only where cheap electrical power is available. However, as will be seen below, the electrolytic method is of special value when the feed water has already been partially enriched in deuterium in other ways.

8.130. The *distillation method* of separating the isotopes of hydrogen is based on a slight difference in the vapor pressures of light and heavy water, as indicated by the small difference in their boiling points. Hence, if water is distilled, the liquid distilling over first contains a slightly higher proportion of the lighter isotope; the residue will thus be somewhat richer in the heavier form. Concentration of heavy water is therefore possible by fractional distillation of natural water. Because of the small difference in the vapor pressures, the distillation columns must consist of a large number of plates or equivalent stages. The greatest separation efficiency is obtained by distillation at reduced pressures. In spite of the magnitude of the equipment required, quantities of heavy water of high purity have been obtained by the fractional distillation of ordinary water. In eight stages of distillation the deuterium content of water has been increased from the normal value of 0.015 per cent to about 90 per cent of the hydrogen present.

8.131. The *chemical-exchange method* makes use of small differences in the reactivities of isotopic molecules. Although isotopes have identical chemical properties, the isotopic forms of a given element or compound frequently react at slightly different rates. As a result the equilibrium constant of an isotopic-exchange reaction, which is the ratio of the specific rates of forward and reverse reactions, differs from unity. This makes possible a partial separation of the isotopes.

8.132. Several hydrogen-deuterium exchange reactions have been studied, and some used, for the production of heavy water. One of these is the reaction between water and hydrogen gas. When the content of deuterium is not too high, this isotope will be present as HD in hydrogen gas and as HDO in water, so that the exchange-reaction equilibrium involving the two isotopes may be written as

$$H_2O + HD \rightleftharpoons HDO + H_2,$$

and the equilibrium constant is

$$K = \frac{(HDO)(H_2)}{(H_2O)(HD)}.$$

If all the substances involved are in the vapor phase, the equilibrium constant ranges from about 3.4 at ordinary temperatures to 2.6 at 100°C (212°F). This means that if hydrogen gas, containing some HD, is allowed to come into equilibrium with water (H_2O) vapor, the net reaction will result in the formation of HDO. The HDO may be scrubbed out of the vapor phase with ordinary water, so that the latter is thereby enriched in deuterium.

8.133. Because the isotopic-exchange equilibrium between water and hydrogen is established slowly, even at 100°C (212°F), it is necessary to use a catalyst; this may be finely divided platinum on a charcoal base or a combination of nickel with chromium sesquioxide. A mixture of water vapor and hydrogen is passed over the catalyst, and the HDO formed is removed by scrubbing with liquid water. The latter is vaporized and again passed over the catalyst together with hydrogen gas, so that more HDO is formed, and so on. If the catalytic exchange reaction and equilibration of the water vapor with the scrubbing water are repeated many times, in a counterflow cascade system, considerable enrichment of deuterium in the water can be achieved.

8.134. The hydrogen-water exchange method requires large volumes of hydrogen gas, and so it is carried out most economically in connection with another process in which hydrogen is an important reactant. The manufacture of ammonia, by synthesis from nitrogen and hydrogen, satisfies these requirements. The hydrogen gas is first fed into the deuterium enrichment plant; here a proportion of the deuterium present is removed, in exchange for the lighter isotope. The residual gas then passes on to the ammonia synthesis, where the slight change in composition is of no significance.

8.135. A modification of the chemical-exchange principle, called the *dual-temperature method*, was proposed several years ago. It makes use of the differ-

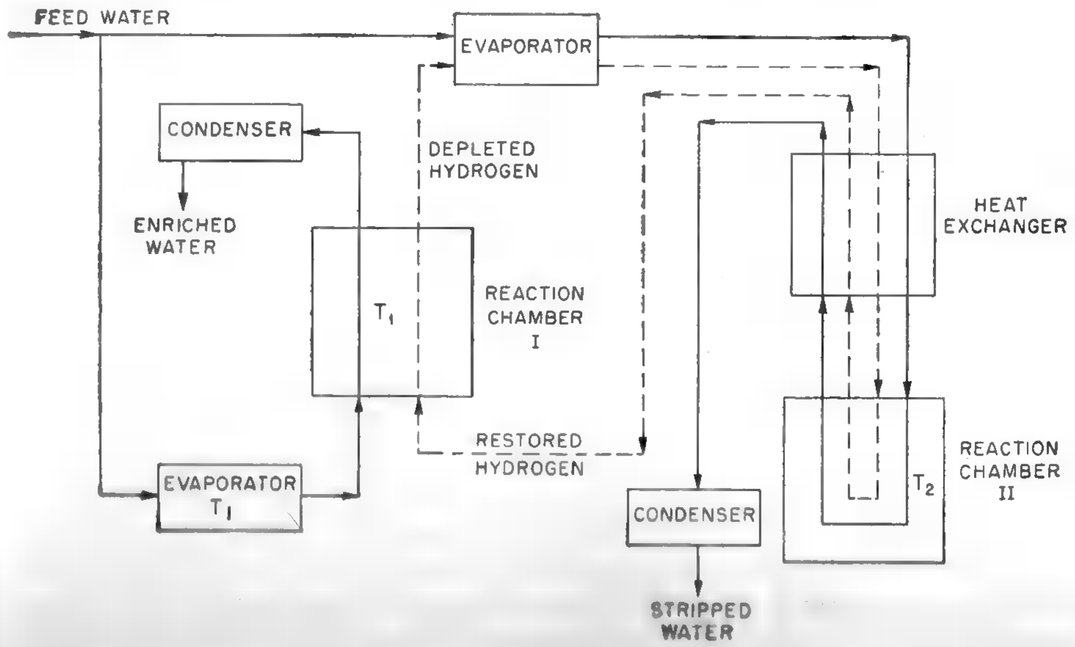


FIG. 8.13. Flow diagram of dual-temperature process for heavy water

ence in the equilibrium constant of an exchange reaction at two temperatures; it does not require large volumes of hydrogen (or other gas) because this gas merely acts as a carrier for deuterium. A simple form of the dual-exchange process, based on the catalytic water-hydrogen exchange reaction considered above, may be illustrated by reference to Fig. 8.13.*

8.136. Feed water, entering at the left, is divided into two parts; one part is evaporated at a fairly low temperature T_1 and then passes into the reaction chamber I where it interacts with hydrogen gas. As a result of the isotopic-exchange reaction the water is enriched in deuterium, whereas the hydrogen is impoverished in this isotope. The other part of the feed water is evaporated at a higher temperature than T_1 and is mixed with the depleted hydrogen gas from chamber I. After passage through a heat exchanger, where its temperature is raised to T_2, which is considerably above T_1, the mixture of water vapor and hydrogen enters the reaction chamber II. Because the equilibrium constant of the water-hydrogen exchange reaction is smaller at T_2 than at T_1 (cf. § 8.132), deuterium is now transferred from the water to the depleted hydrogen. After removal of the water by condensation, the hydrogen, with its restored deuterium content, returns to the low-temperature reaction chamber I where it is once more depleted, and so on. The unit shown in Fig. 8.13 may be regarded as one dual-temperature stage in a cascade, in which the water is steadily enriched in deuterium as it passes from one stage to the next.

8.137. In considering the problem of concentrating heavy water from 0.015 per cent to about 99.75 per cent, which is desirable for use in reactors, it appears that no one process is superior to the others over the whole range of concentrations. Most of the cost of the enrichment operation is incurred in the earliest stages, mainly because large plants are necessary to handle the enormous volumes of water which must be treated. As the proportion of heavy water increases, however, the volumes become much less and the situation is changed. One of the consequences is that electrolysis, which is generally prohibitively expensive for the early stages, can well be used for the later stages of enrichment. The most economical approach is, therefore, to use either distillation or chemical exchange, or both, to produce some increase in the deuterium content of ordinary water and then to bring the product up to the desired concentration by electrolysis.

8.138. Brief reference may be made, in conclusion, to the possibility of separating deuterium from hydrogen by fractional distillation of liquid hydrogen at low temperatures. In the opinion of some competent authorities, this method should be capable of producing heavy water at a lower cost than any of the processes described above. Although the boiling points of liquid hydrogen and liquid deuterium at atmospheric pressure differ by only 3.12°C (5.6°F), the ratio of their vapor pressures, at a constant temperature, is exceptionally large. Thus, at about 22°K (40°R), which is between the boiling points of the pure liquids,

* E. Cerrari, *et al.*, *Chem. Eng. Prog. Symposium Series*, No. 11, **50**, 271 (1954).

the ratio of the vapor pressures for H_2/HD is 1.6 and for H_2/D_2 it is nearly 2.5. This high relative volatility means that efficient separation is possible by fractional distillation, with regard both to the number of theoretical stages required and to the extent of recovery of deuterium.

8.139. The main problems associated with the process are (1) the operation of a large-scale plant at temperatures in the vicinity of −250°C (−420°F) at which the liquefaction and rectification of the hydrogen would have to be carried out; (2) the formation of solid deposits in the feed lines at these low temperatures due to the presence of traces of impurities in the hydrogen; and (3) the danger associated with the handling of large quantities of liquid hydrogen. Although detailed designs for a plant for the separation of deuterium by the fractional distillation of liquid hydrogen have been prepared, there is as yet no report that such a plant has been constructed.

DECOMPOSITION OF WATER BY NUCLEAR RADIATIONS

Experimental Observations

8.140. Because water, either in the ordinary form or as heavy water, is used as moderator, solvent, or coolant in a number of reactors, the action of nuclear radiations is of particular interest. The discussion given below usually applies equally to all isotopic forms of water, i.e., H_2O, HDO, or D_2O, or to mixtures; wherever reference is made to "water" or to "hydrogen" any of the isotopic forms may be implied according to circumstances.

8.141. Upon exposure of water to nuclear radiations, hydrogen and oxygen gases are liberated, and hydrogen peroxide can be detected in the aqueous phase. If the water is contained in a closed system, the gas pressure increases until a steady-state or equilibrium value is attained. The magnitude of this steady-state pressure depends on the nature of the radiation, its density (or flux), the temperature, and the presence of various dissolved substances in the water. In general, the steady-state gas pressure increases with the radiation flux but decreases with increasing temperature. Many dissolved impurities also bring about an increase in the steady-state pressure. The concentration of hydrogen peroxide in the solution is decreased by raising the temperature and by the presence of impurities.

8.142. It may be noted that slow neutrons probably do not cause direct decomposition of water. However, as a result of radiative capture by (ordinary) hydrogen atoms, gamma rays of 2.21 Mev energy are produced, and these are capable of decomposing the water.† The effect of fast neutrons is probably to

* W. J. Toulis, Report UCRL-583; A. O. Allen, Report AECU-1413; *J. Phys. Chem.*, **56**, 575 (1952); *Chem. Eng. Prog. Symposium Series*, No. 12, **50**, 238 (1954); C. J. Hochanadel, *J. Phys. Chem.*, **56**, 587 (1952).

† This secondary decomposition of water due to slow neutrons would be much less marked in heavy water because of the low neutron capture cross section of deuterium.

be attributed to the high-energy recoil protons produced by the collision of the neutrons with hydrogen nuclei.

Interpretation of Experimental Data

8.143. In seeking to interpret the foregoing results, it may be postulated, in the first place, that ionizing radiation causes the decomposition of water into hydrogen atoms and free hydroxyl radicals; thus,

$$H_2O \rightsquigarrow H + OH.$$

This decomposition probably does not occur directly, but the intermediate stages are of no significance here and so will be omitted from consideration. Subsequently the H and OH may either recombine to form water, i.e.,

$$H + OH = H_2O,$$

in which case there is no net decomposition, or they may combine in pairs, i.e.,

$$H + H = H_2$$

and

$$OH + OH = H_2O_2,$$

to yield the observed products, hydrogen and hydrogen peroxide, respectively. If the specific ionization of the radiation is low, as it is with gamma rays, the H and OH formed by the decomposition of a single water molecule will be much closer together than the H and H (or OH and OH) from two different water molecules. Consequently the tendency for recombination to occur will be greater than for radiation having a high specific ionization.

8.144. Since hydrogen peroxide is not very stable, it will decompose to some extent according to the reaction

$$H_2O_2 = H_2O + \tfrac{1}{2}O_2,$$

so that oxygen gas is liberated. It should be noted that, as long as some hydrogen peroxide remains undecomposed, the ratio of hydrogen to oxygen gases formed will be more than 2 to 1, the atomic ratio in water. As the temperature is raised, the decomposition of hydrogen peroxide will be favored, and more oxygen will be liberated. This expectation is in accord with experiments which show that at high temperatures, e.g., 250°C (480°F), the irradiated aqueous systems contain little or no hydrogen peroxide and that hydrogen and oxygen are present in the evolved gas in a proportion close to 2 to 1. Impurities which catalyze the decomposition of hydrogen peroxide have the same general effect.

8.145. In view of the occurrence of a steady-state gas pressure in a closed system, it is apparent that there must occur a reverse reaction which tends to remove molecular hydrogen. It has been suggested that this takes place through a chain mechanism, the first stage of which is the interaction between hydrogen atoms and molecules of hydrogen peroxide, i.e.,

$$H + H_2O_2 = H_2O + OH.$$

This is followed by reaction between the hydroxyl radicals and molecular hydrogen, i.e.,

$$OH + H_2 = H_2O + H,$$

so that hydrogen atoms are regenerated and can carry on the chain. The net result, obtained by adding the two equations, is

$$H_2 + H_2O_2 = 2H_2O,$$

which means that the molecular hydrogen is removed.

8.146. The actual pressure attained at the steady state will represent a balance between the rate of gas formation by the action of radiation and its removal by the chain reaction involving hydrogen atoms and hydroxyl radicals. Increase in the radiation density will obviously be expected to increase the steady-state pressure. On the other hand, impurities present in solution may react with hydrogen atoms and hydroxyl radicals and may destroy them. As a result the reverse (chain) reaction, i.e., removal of molecular hydrogen, is inhibited, and the steady-state pressure will be increased.

8.147. The explanation of the effect of increased temperature in decreasing the steady-state gas pressure is based on the supposition that hydrogen peroxide, in addition to taking part in the chain reaction which removes molecular hydrogen, is also involved in a chain reaction which destroys hydrogen atoms and hydroxyl radicals, namely,

$$H_2O_2 + OH = HO_2 + H_2O$$
$$HO_2 + H = H_2O_2,$$

the net effect being

$$H + OH = H_2O.$$

Since increase of temperature results in the decomposition of hydrogen peroxide, the destruction of hydrogen atoms and hydroxyl radicals is reduced. If this particular chain is inhibited to a greater extent than the one considered in § 8.145, leading to the removal of molecular hydrogen, the over-all effect will be a decrease in the steady-state gas pressure.

Hydrogen-Oxygen Gas Recombination

8.148. It is inevitable that in any reactor using either ordinary or heavy water as moderator, reflector, or coolant there should be some decomposition into hydrogen (or deuterium) and oxygen due to the action of various radiations. Provision must, therefore, be made for the removal or recombination of the gases in this explosive mixture. The problem is aggravated in homogeneous reactors, consisting of a uranium salt dissolved in water, because the fission products are formed within the solution. Since these products have high ener-

gies, they cause considerable decomposition of the water. From published information relating to the Los Alamos Water Boiler* and the HRE at Oak Ridge† it appears that, in spite of the different characteristics of these reactors, in each case the hydrogen-oxygen mixture is liberated at the rate of about 1 ft^3 (measured at normal temperature and pressure) per min per 100 kw of reactor power.

8.149. In order to dispose of the gases formed by the radiation decomposition of water in these reactors, recombining systems are used. In the Los Alamos and Raleigh (North Carolina State College) Water Boilers a catalytic method for combining hydrogen and oxygen is employed. The gases liberated during reactor operation are first passed through a condenser and an entrainment filter, in which most of the moisture is removed, and then on to a catalyst chamber. The catalyst consists of small alumina pellets coated with finely divided platinum. Combination of the hydrogen and oxygen occurs and the heat released in the reaction raises the temperature sufficiently to convert the water formed into steam. The steam is condensed and the resulting water is returned to the reactor.

8.150. When the HRE operates at high power the hydrogen-oxygen gas mixture from the reactor, after removal of moisture, enters a flame recombiner. This is a device similar to a large Bunsen-type burner in which the hydrogen actually burns as a flame while combining with oxygen. The exit gases from the flame recombiner may still contain small amounts of hydrogen and oxygen, and these are removed by means of a platinum-alumina catalyst similar to that described above. The steam formed in both recombiners is condensed and the water is returned to the reactor. When operating at low powers the use of the flame recombiner is not necessary and the reactor gases pass directly to the catalyst chamber.‡

8.151. It is of interest to note that, on the basis of the data given in § 8.148, concerning the rate of evolution of the hydrogen-oxygen gas mixture, it can be calculated that about 4 per cent of the reactor energy appears as heat in the recombining system. If a homogeneous reactor were designed for industrial power production, provision would, of course, be made for the use of this heat.

* L. D. P. King, *et al.*, *Nucleonics*, **11**, No. 9, 25 (1953).

† S. E. Beall and C. E. Winters, *Chem. Eng. Prog.*, **50**, 256 (1954).

‡ The reactor gases contain small amounts of highly radioactive xenon and krypton which are unaffected by the recombining systems. These substances are either adsorbed on activated carbon and buried, or, if in sufficiently small amounts, they are mixed with large volumes of air and discharged to the atmosphere through a stack after a suitable delay time (cf. H. M. Busey, *Nucleonics*, **12**, No. 5, 9 (1954)).

BERYLLIUM AS MODERATOR AND REFLECTOR *

General Properties

8.152. Although the nuclear properties of beryllium are such as to make it a valuable material for use as moderator or reflector in a thermal reactor, with the added advantage of being a good thermal conductor, it has hitherto been used only to a limited extent. In the MTR, metallic beryllium is employed in the reflector, and the plans for the Submarine Intermediate Reactor (SIR) call for this element to serve as moderator. The use of beryllium or its oxide or carbide has also been proposed for other reactors that are now either in the experimental or in the design stage.

8.153. The reflector of the original (LOPO) form of the Water Boiler consisted mainly of beryllium oxide, but this was later replaced with graphite. The chief reason was that the photoneutrons, which continued to be formed by the (γ, n) reaction with beryllium after the reactor was shut down, affected the start-up routine. In general, however, photoneutrons, which are also produced from heavy water, might be regarded as an advantage, since they would represent an appreciable and detectable source, provided the reactor has not been shut down too long (see § 6.164).

8.154. Hitherto, beryllium of sufficient purity for reactor use has been expensive; in addition, it was reported to be brittle, difficult to fabricate, and very susceptible to corrosion in water, especially at high temperatures. With the development of new techniques in powder metallurgy and the availability of purer materials, metal of greatly improved properties with regard to fabrication, mechanical characteristics, and corrosion resistance has been prepared. Progress has also been made in the production and fabrication of the oxide and carbide. It is possible, therefore, that beryllium may play an increasingly significant role in reactor design. Mention may be made here of the fact that beryllium and its compounds are a potential health hazard. Special precautions must be taken to avoid inhalation or ingestion of beryllium in any form.

Production of Beryllium

8.155. Although several beryllium minerals are known, the only commercial source of the element at the present time is *beryl*, $3BeO \cdot Al_2O_3 \cdot 6SiO_2$. The theoretical beryllium content is 5.0 per cent, but ores containing 3 per cent or more of the element (8 to 14 per cent of BeO) are acceptable raw materials. Most of the beryl now being used in the United States originates in Brazil, Argentina, and India, but increasing quantities of the ore are coming from other

* For reviews, see M. C. Udy, H. L. Shaw, and F. W. Boulger, *Nucleonics*, **11**, No. 5, 52 (1953); H. A. Saller, *Proc. Conf. on Nuc. Eng.*, Berkeley, 1953, p. 11; W. A. Johnson, *Chem. Eng. News*, **32**, 1882 (1954); E. J. Boyle and J. L. Gregg, *Chem. Eng. Prog. Symposium Series*, No. 11, **50**, 53 (1954); D. W. White, Jr., and J. E. Burke (Eds.), "The Metal Beryllium," American Society for Metals, 1955; C. A. Hampel (Ed.), "Rare Metals Handbook," Reinhold Publishing Corp., 1954, Chapter 3 (by B. R. F. Kjellgren).

sources. Substantial amounts of domestic beryl are also available, mainly as a by-product of some other operation, such as feldspar mining. If the high-grade ores now being worked become exhausted, it is believed that all reasonable requirements can be met by ores of lower grade.

8.156. In outline the method for producing metallic beryllium from beryl, of which a simplified flow sheet is given in Fig. 8.14, is somewhat as follows: The ore is first heat-treated and ground to render it attackable by sulfuric acid. The latter converts the beryllium and aluminum, as well as the iron and alkali metals present as impurities, into soluble sulfates, leaving a residue of silica. Ammonium hydroxide is added to form ammonium sulfate with the excess sulfuric acid, and the bulk of the aluminum is crystallized out as ammonium

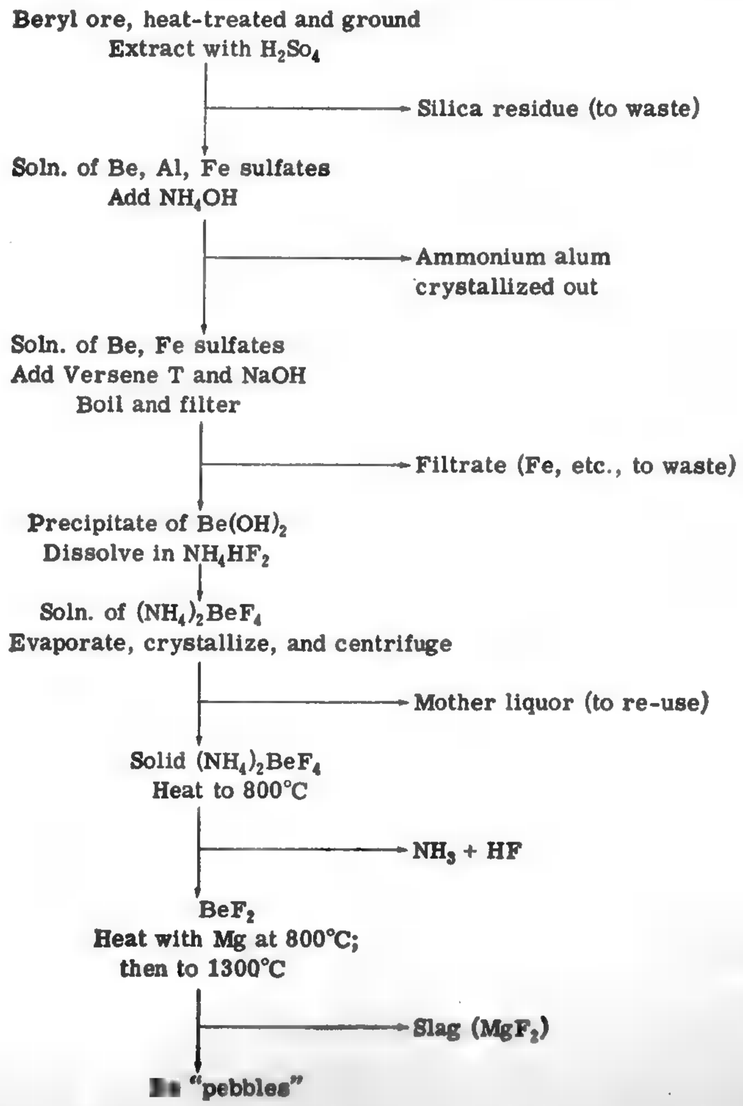


FIG. 8.14. Simplified flow sheet for extraction of beryllium from beryl

aluminum sulfate (ammonium alum). The residual solution is treated with the sodium salt of ethylenediamine tetraacetic acid (Versene T) which "sequesters" the iron as a complex molecule, thus keeping it in solution, when excess of sodium hydroxide is added to convert the beryllium into soluble sodium beryllate, Na_2BeO_4. Upon boiling, the latter decomposes and precipitates beryllium hydroxide, but the iron remains in solution.

8.157. The precipitate of beryllium hydroxide is dissolved in aqueous ammonium acid fluoride (NH_4HF_2), forming a solution of ammonium beryllium fluoride, $(NH_4)_2BeF_4$. This solution is evaporated, and the double salt which crystallizes out is separated by centrifuging. The solid is decomposed by heating to 800°C (1470°F), leaving a residue of beryllium fluoride. It is at this stage that most of the boron present as impurity is removed, presumably as the volatile boron trifluoride. The product contains less than 1 ppm of boron. The reduction of beryllium fluoride to the metal is carried out by means of magnesium at 800°C (1470°F) in graphite crucibles. The beryllium is first formed as a finely divided powder, but upon heating to 1300°C (2370°F) the particles coalesce into pockets of liquid. After cooling and removal of the slag, the beryllium is obtained in the so-called "pebble" form.

8.158. The pebbles, which contain from 96 to 99 per cent of beryllium, are cleaned from slag, melted in a vacuum furnace in beryllium oxide crucibles, and cast into ingots. This process reduces the magnesium content of the material and also removes slag particles that may have been occluded in the pebbles. The beryllium content of the vacuum-cast ingots is over 99 per cent, but a small proportion of the oxide is almost invariably present.

8.159. If beryllium oxide is required, the beryllium hydroxide is dissolved in sulfuric acid. Beryllium sulfate is crystallized from the solution and is heated strongly to produce beryllium oxide in a fluffy form.

8.160. For the production of beryllium carbide, beryllium powder is mixed with the required proportion of pure graphite and allowed to react in a hot-pressing or sintering furnace. The reaction temperature depends on the particle size, but is usually about 900 to 1000°C (1650 to 1830°F). The formation of beryllium carbide takes place readily under mechanical pressure.

PHYSICAL AND MECHANICAL PROPERTIES OF BERYLLIUM

8.161. Beryllium is a steel-gray metal with a melting point of 1280°C (2340°F). The mechanical properties of beryllium can be changed considerably by working and by metallurgical treatment, but, in general, it is more brittle than other metals of high purity. Only one crystalline form, with a hexagonal close-packed structure, has been observed between ordinary temperatures and the melting point. The lattice constants are $a_0 = 2.281$ A and $c_0 = 3.577$ A, and the theoretical density is 1.848 g/cm³ at 25°C (78°F). The observed densities of different specimens of beryllium range from 1.81 to 1.86 g/cm³, the higher values being due to the presence of beryllium oxide, theoretical density 3.0 g/cm³. Because

of the hexagonal close-packed structure, many of the physical and mechanical properties of the metal are anisotropic in nature. However, material made by hot-pressing beryllium powder is isotropic, presumably because of the random orientation of the small grains. At ordinary temperature the coefficient of thermal expansion is about 12×10^{-6} per °C. The thermal conductivity is 0.35 cal/(sec)(cm²)(°C/cm) at 25°C (78°F), decreasing to 0.23 at 600°C (1110°F). The electrical resistivity is approximately 4×10^{-6} ohm-cm at room temperatures.

8.162. The mechanical properties of specimens of beryllium metal vary with the methods of fabrication and treatment. Since hot-pressing of the powder has proved a very successful fabrication procedure, as will be seen below, the properties recorded in Table 8.11 refer to material obtained by hot-pressing in

TABLE 8.11. MECHANICAL PROPERTIES OF HOT-PRESSED BERYLLIUM

Ultimate tensile strength, psi	45×10^3
Yield strength (0.2% offset), psi	32×10^3
Elongation in 2 in., %	2.3
Modulus of elasticity, psi	44×10^6
Poisson's ratio	0.024
Unnotched Charpy impact, ft-lb	0.8
Brinell number	120–150

vacuum at a temperature of about 1000°C (1830°F).* The small elongation at tensile fracture and the low impact resistance even for unnotched bars indicate that the ductility of the material is relatively low. The ductility of cast beryllium is, in general, even lower. By warm-extruding the hot-pressed metal and then annealing at about 700°C (1290°F), the mechanical properties, especially in the direction of extrusion, are greatly improved. The brittle nature of beryllium has been partly attributed to the presence of impurities, but this is not certain. Metal of a very high degree of purity is now becoming available, and measurements made upon it may throw some light on the problem.

Fabrication of Beryllium

8.163. Castings of beryllium have been made by melting the metal in a beryllium oxide crucible and pouring into graphite molds, but cracks may develop during or immediately after solidification unless the rate of cooling is carefully controlled. The cast material does not fabricate easily; because of the large grain size it is difficult to machine without causing surface damage, and cracking may occur below the surface. Although cold-working is not practical, cast beryllium can be hot-worked to some extent by forging and rolling, provided it is encased in a more malleable metal such as steel. Cast billets can also be

* W. W. Beaver and K. G. Wikle, *J. Metals*, **6**, 559 (1954).

extruded at high temperatures with the use of a steel jacket. Pieces of beryllium can be joined by welding or brazing, but the technique is complicated by the tendency for beryllium to oxidize in air.

8.164. The development of powder-metallurgical processes has represented an important advance in the fabrication of beryllium.* It has been reported that most billets for further fabrication are now produced by hot-pressing of the powder in vacuum. The ingots of vacuum-cast beryllium are first chipped and then reduced to fine powder in an attritioning mill in the absence of air. The powder is then compacted into the desired shape under vacuum at temperatures in the vicinity of 1000°C (1830°F) and pressures of about 500 psi. Pressings with good mechanical properties (see Table 8.11), weighing up to several hundred pounds, have been produced in this manner. The hot-pressed material can be machined fairly easily and long holes of small diameter can be drilled in it.

8.165. Although the hot-pressed beryllium is frequently used without any further treatment, other than machining to size, it can be hot-extruded at 800 to 1100°C (1470 to 2010°F) or warm-extruded at between 300 to 500°C (570 to 930°F). As stated earlier, the resulting products exhibit an increase of ductility in the direction of extrusion.

Corrosion of Beryllium

8.166. It was thought at one time that beryllium metal had poor corrosion resistance in water, but the effects observed were probably due to the presence of localized impurities. By proper treatment, specimens of beryllium can be obtained which withstand the action of deaerated distilled water at 250°C (480°F) for many days. In the presence of oxygen (or air) the corrosion is increased.

8.167. Fresh surfaces of beryllium metal soon tarnish upon exposure to air, indicating the formation of a thin oxide film. This film, however, retards further oxidation. Consequently, in massive form, beryllium does not suffer serious attack in air below 600°C (1110°F), although the powdered metal is oxidized at lower temperatures. Above 600°C (1110°F) the oxide is first formed, and then the nitride (Be_3N_2) at about 1000°C (1830°F). The use of beryllium at temperatures in excess of 600°C (1110°F) in the presence of air is thus not recommended. It is of interest to mention that at high temperatures beryllium is much less readily attacked by oxygen than is titanium or zirconium.

8.168. No appreciable change was observed after exposure of beryllium to liquid bismuth, lead, or tin at 700°C (1290°F) for 24 hr. Liquid gallium, which had first been saturated with beryllium (about 1 per cent), showed little attack at 500°C (930°F). At 600°C (1110°F) and above, there was evidence of grain-boundary type of erosion.

* See A. U. Seybolt, Report AECU-1029; H. H. Hausner and N. P. Pinto, *Trans. Am. Soc. Metals*, **43**, 1092 (1951); N. P. Pinto, *J. Metals*, **6**, 629 (1954); H. H. Hausner and H. B. Michaelson, *Chem. Eng. Prog. Symposium Series*, No. 11, **50**, 11 (1954).

GRAPHITE AS MODERATOR AND REFLECTOR*

Production of Graphite

8.169. Graphite has probably been used more extensively than any other moderator or reflector in existing reactors. Its nuclear properties are not quite as good as those of heavy water or beryllium, but graphite of satisfactory purity is readily available at reasonable cost. It has good mechanical properties and thermal stability. However, at high temperatures it reacts with oxygen (in air) and with water vapor, and with some metals and metal oxides it forms carbides. Although it is a nonmetal, graphite is a good conductor of heat, a desirable property for a moderator. Its chief drawbacks under normal operating conditions are the possibility of oxidation in the presence of air, relatively low impact strength, and the porous nature of the material used in reactors. As stated earlier, it is also affected by nuclear radiations.

8.170. Graphite occurs in considerable quantities in nature, but, since in this state it is relatively impure, the reactor-grade product is made artificially by "graphitization" of petroleum coke. The coke is first heated to drive off volatile matter and is then ground and mixed with a coal-tar pitch binder at a temperature suitable for fluidizing the latter. The mixture is extruded into bars and baked, in gas-fired ovens, to temperatures up to 1500°C (2730°F), in order to carbonize the pitch and set the binder. To increase the bulk density of the product, it is impregnated with pitch under vacuum and again baked. The resultant gas-baked carbon is finally graphitized by electrical-resistance heating. The temperature is raised to about 2700 to 3000°C (4860 to 5430°F) during the first few days and then about three or four weeks are allowed for the material to cool. The physical characteristics of the final product vary to some extent with the fineness of the ground coke, the type and amount of pitch used for impregnation, and the temperature and duration of the graphitization treatment.

Physical and Mechanical Properties of Graphite†

8.171. Graphite is a black, moderately soft, allotropic form of carbon; it is the stable phase at all reasonable temperatures. It does not melt upon heating at atmospheric pressure but sublimes at about 3650°C (6600°F). The crystals have hexagonal symmetry and consist of flat layers or planes of carbon atoms, stacked parallel to one another.‡ The planes are relatively far apart, namely, 3.41 A, so that shear occurs easily by a slipping motion of the planes relative to each other. This type of shear, perpendicular to the hexagonal axis, accounts for the softness

* S. G. Bauer, *Research*, **5**, 515 (1952); L. D. Loch and J. A. Slyh, *Chem. Eng. Prog. Symposium Series*, No. 11, **50**, 39 (1954).

† L. D. Loch and J. A. Slyh, *loc. cit.*; C. Malmstrom, R. Keen, and L. Green, *J. Appl. Phys.*, **22**, 593 (1951); J. P. Howe, *J. Am. Ceram. Soc.*, **11**, 275 (1952).

‡ Because of small discrepancies in the bond angles, graphite is sometimes regarded as orthorhombic; however, the departure from hexagonal symmetry is very slight.

and lubricating properties of flake graphite. As is to be expected, the thermal, electrical, and mechanical properties of graphite crystals are anisotropic, and this behavior is exhibited to some extent by the polycrystalline material because of preferred orientation of the individual crystallites.

8.172. The physical and mechanical properties of graphite vary widely among different products. The theoretical density, calculated from the lattice constants, is 2.27 g/cm³, and this value is approached by natural graphite. However, the material manufactured for reactors has a density of 1.6 to 1.7 g/cm³; this low density is due to the porosity of the artificial product.*

8.173. The values of a few important properties of graphite at ordinary temperatures are recorded in Table 8.12. Because of the considerable variations

TABLE 8.12. APPROXIMATE VALUES OF PROPERTIES OF GRAPHITE

Coefficient of thermal expansion, per °C	2×10^{-6} (long.) 3×10^{-6} (trans.)
Thermal conductivity, cal/(sec)(cm²)(°C/cm)	0.4
Tensile strength, psi	3000
Compressive strength, psi	3500
Modulus of elasticity, psi	1.2×10^6

that have been observed among different samples, the data are to be regarded as approximate only; their purpose is merely to provide a rough qualitative indication of some of the physical and mechanical properties of graphite at normal temperatures. As indicated above, the values may vary with direction. The tensile strength of graphite increases with temperature, attaining a maximum at about 2500°C (4530°F), when the value is roughly twice that at ordinary temperatures. The creep rate is very small below 1500°C (2730°F), but may become appreciable at higher temperatures. Graphite has excellent resistance to thermal shock, and it is difficult to produce fracture by thermal stress alone.

REACTOR STRUCTURAL MATERIALS†

General Characteristics

8.174. Every reactor system must inevitably include a certain amount of structural material which serves as a mechanical framework for the reactor components. In addition, various types of containers are required for solid or liquid fuels, for coolants, for control rods, and for measuring instruments. Because uranium is readily attacked by air, water, and other fluids which might be used as coolants, it is necessary that fuel elements be protected with suitable cladding. Such cladding is necessary, in any event, to prevent escape of fission products from the fuel element.

* Reactor-grade graphite with a density ~2 g/cm³ has been reported from West Germany.

† For reviews see J. E. Burke, Report AECU-2321; W. V. Koshuba and V. P. Calkins, *Metal Prog.*, **62**, No. 1, 67 (1952); G. E. Evans, *Nucleonics*, **11**, No. 6, 18 (1953).

8.175. The requirements of a structural material will vary to some extent with the type of reactor and with its specific purpose in the reactor. There are, however, some general characteristics which may be enumerated. Mechanical properties, such as tensile strength, impact strength, and rupture stress, must be adequate for the operating conditions. The material must be capable of being fabricated or joined into the required shapes. Thermal conductivity should usually be high and the coefficient of thermal expansion low or well matched with that of other materials.

8.176. In many reactor components there is very considerable internal heating, due either to fission, to the slowing down of fast neutrons, or to the absorption of various radiations. The removal of the heat from the exterior results in high temperature gradients within the material. Such materials must, therefore, be able to maintain stability under severe thermal stress.

8.177. Corrosion and erosion problems in a reactor are often unusually serious. Corrosion may arise from the galvanic action between two different metals or a metal and a nonmetallic conductor, e.g., graphite, when in contact in an aqueous medium or a fused salt. Even in a single metal, stresses, strains, and other defects, produced during fabrication, or the presence of oxide or other inclusions can facilitate corrosion. When there are temperature differences in a circulating system, e.g., of liquid-metal coolant, especially at high temperatures, corrosion of the mass-transfer type may occur. Metal is then dissolved by the circulating liquid in regions where the temperature is high and is deposited in the cooler parts of the system.*

8.178. In addition to the physical and mechanical properties referred to above, the nuclear properties must be satisfactory. If the material is to be used in or near the core or reflector of the reactor, it must obviously have a small cross section for neutron absorption. It should be noted in this connection that a material that cannot be used in a thermal reactor because of a large cross section may perhaps be employed in a fast reactor since the cross section for the absorption of fast neutrons may then be tolerable.

8.179. As a result of neutron capture, many materials become radioactive and consequently are dangerous to handle. Thus the maintenance or repair of equipment exposed to the neutron flux of a reactor may present a difficult problem. In the choice of materials, preference should be given, if possible, to substances which either do not become radioactive as a result of neutron capture or for which the induced activity is weak, with no gamma radiation, or of short half life. In this connection, the data in Table 8.13 will be of some value. First there are listed the elements which might have some interest for structural purposes or which might be present as alloying elements or as impurities. In each case the mass number is given of the isotope (or isotopes) which yields a

* Experience with mercury has shown that mass transfer in the flowing liquid (due to a temperature gradient) can be controlled by the addition of small amounts of other metals which act as inhibitors.

TABLE 8.13. INDUCED ACTIVITIES IN METALS PRESENT IN STRUCTURAL MATERIALS

Element	Isotope Mass No.	Natural Abundance (per cent)	σ_a (barns)	Active Species	Half Life	γ-Ray Energy (Mev)
Titanium	50	5.3	0.04	Ti^{51}	72d	1.0
Chromium	50	4.4	16	Cr^{51}	27d	0.32
Manganese	55	100	13.3	Mn^{56}	2.6h	2.1
Iron	58	0.33	0.8	Fe^{59}	46d	1.3
Cobalt	59	100	37	Co^{60}	5.3y	1.3
Nickel	64	1.9	3.0	Ni^{65}	2.5h	0.93
Copper	63	69	4.3	Cu^{64}	12.8h	1.35
Zinc	64	48.9	0.5	Zn^{65}	250d	1.12
	68	18.5	0.1	Zn^{69}	13.8h	0.4
Zirconium	94	17.4	0.1	Zr^{95}	65d	0.92
Molybdenum	98	23.8	0.13	Mo^{99}	67h	0.84
Tantalum	181	100	21.3	Ta^{182}	113d	1.2
Tungsten	186	28.4	34	W^{187}	24h	0.76

radioactive species as a result of neutron absorption, its abundance in the element as it occurs in nature, and its thermal absorption cross section. The next column gives the identity of the active species, and this is followed by its half life and the energy of the most penetrating (highest energy) gamma rays. It should be emphasized that the information in the table refers only to radioactive isotopes of appreciable half life (>1 hr) emitting gamma radiations of moderate or high energy (>0.3 Mev), in addition to beta particles. There are other species which are beta-active only, but these do not usually present a serious maintenance problem, provided the bremsstrahlung are avoided.

8.180. The elements which may be regarded as making the major contribution to persistent induced activity are chromium, manganese, cobalt, copper, zinc, tantalum, and tungsten. It will be seen shortly that none of these is likely to be used as a thermal reactor constituent in a moderately pure form. However, these metals may well be present as impurities or as alloying elements in otherwise acceptable structural materials, and consideration should be given to the activity that will result from exposure to neutrons.

Choice of Structural Materials

8.181. The crucial requirement for a material to be used in a thermal reactor is that it shall have a reasonably low absorption cross section for neutrons. If the cross section is large, the material must be rejected no matter what mechanical or other advantages it may possess. If there are compensating factors, an element with a moderate cross section may be tolerated, but its presence will require the use of additional fissionable material in the reactor core. In a thermal-neutron breeder reactor, where every neutron is significant, parasitic

capture would have to be avoided if at all possible. Since the capture cross sections of nearly all elements are small for neutrons of high energy, the choice of materials for fast-neutron reactors is much less restricted.

8.182. In order to obtain an over-all picture of the possibilities with regard to materials for thermal reactors, metallic elements can be divided into three classes: (1) elements with low capture cross sections for thermal neutrons, i.e., below 1 barn; (2) elements with cross sections of intermediate value, i.e., from 1 to 10 barns; and (3) elements with cross sections exceeding 10 barns. In general, those in the last category, which include tungsten, tantalum, and cobalt, need not be considered further, except perhaps for intermediate and fast reactors. Of the substances having small or moderate cross sections, soft metals and those with melting points below 500°C (930°F) may be eliminated as not likely to be very useful for structural purposes. The thermal absorption cross sections and melting points of the remaining elements, which are more or less readily available, are given in Table 8.14.

TABLE 8.14. PROPERTIES OF POSSIBLE STRUCTURAL MATERIALS

Low Thermal Cross Sections				Intermediate Thermal Cross Sections			
		M.P.				M.P.	
Metal	σ_a, barns	(°C)	(°F)	Metal	σ_a, barns	(°C)	(°F)
Beryllium	0.009	1280	2340	Columbium	1.1	2415	4380
Magnesium	0.069	651	1200	Iron	2.4	1539	2804
Zirconium	0.18	1845	3350	Molybdenum	2.4	2625	4760
Aluminum	0.22	660	1215	Chromium	2.9	1850	3360
				Copper	3.6	1083	1981
				Nickel	4.5	1455	2651
				Vanadium	5.1	1900	3450
				Titanium	5.6	1670	3040

8.183. The number of metals with small thermal-neutron cross sections is seen to be very limited. Aluminum, usually in the relatively pure (>99.0 per cent) 2S form, has been extensively used as a reactor structural material, for cladding fuel elements, and for other purposes not involving exposure to high temperatures. Although magnesium has a slightly smaller cross section than aluminum, it is doubtful whether this would offset its higher cost and the problems associated with its fabrication. The same is true, to a large extent, for beryllium, although this metal might be used as a structural material in such circumstances that advantage could also be taken of its value as a neutron moderator and reflector.

8.184. The remaining metal, zirconium, has only recently become available in commercial quantities of reasonable purity. It appears to be one of the most promising structural and cladding materials for nuclear reactors, especially those employing water at high pressure (and temperature) as the coolant. In addition to its low capture cross section for thermal neutrons, zirconium has excellent mechanical and fabrication properties and is exceptionally corrosion-resistant. Its high melting point is somewhat misleading, however, as it suffers a decrease in mechanical strength, and also becomes less resistant to attack, at temperatures above about 400°C (750°F). The production and properties of zirconium will be considered more fully later (§ 8.191).

8.185. Turning to the elements of intermediate cross section, it will be seen that here also the choice is very limited. Columbium (niobium) has good mechanical properties but it is scarce and oxidizes badly above 200°C (390°F). The difficulty of fabricating chromium and molybdenum and the relatively high cost of vanadium make it unlikely that these metals will serve any purpose other than as alloying elements. On the other hand, the properties of iron, nickel, and copper can be so greatly improved by suitable alloy additions that their use in the unalloyed form is not probable.

8.186. The other metal in this category, titanium, has attracted much attention in recent years.* Titanium ores are abundant and are found concentrated in large deposits, chiefly as the minerals *rutile*, TiO_2, and *ilmenite*, $FeTiO_3$. These compounds are difficult to reduce to the metal, but commercial processes have been developed and are being improved. The special interest in titanium has resulted largely from its high strength-to-weight ratio in the temperature range from 100 to 450°C (212 to 842°F), which makes it attractive for uses where weight-saving is important. The corrosion resistance of the metal in certain environments is outstanding, and it holds considerable promise for use with aqueous solutions at high temperatures. The fabrication of parts from titanium by forging and welding presents some problems, but continuous progress is being made in overcoming them. The relatively high cross section will limit its use in reactor cores, but its resistance to corrosion may lead to applications in those parts of reactor systems where neutron absorption is not critical, e.g., for the transfer of solutions in aqueous homogeneous reactors and their associated chemical processing equipment.

Alloys as Structural Materials

8.187. In addition to the pure metals considered above, there are many possibilities of producing alloys with desirable properties for use in reactor construction. Besides the various combinations among the metals listed in Table 8.14, such elements as silicon (σ_a = 0.1 barn) and tin (σ_a = 0.6 barn), which would not normally be regarded as structural materials, might prove useful for alloying

* C. A. Hampel (Ed.), "Rare Metals Handbook," Reinhold Publishing Corp., 1954, Chapter 24 (by H. R. Ogden and R. W. Gonser).

purposes. With the increasing availability of pure beryllium, zirconium, and titanium, it is possible that new alloys may be developed of special interest for reactor applications.

8.188. Among the more familiar materials, the austenitic stainless steels are corrosion-resistant and have good mechanical properties. The factors governing their selection for reactors and the problems connected with their use are similar to those encountered in other chemical and high-temperature applications. In spite of their generally satisfactory performance in a wide variety of circumstances, the choice of an austenitic stainless steel can be made only after a thorough investigation of the potential difficulties which may develop in its use. Among the problems to be considered are the following: (1) susceptibility, in certain environments, to intergranular corrosion of unstabilized stainless steels after sensitizing heat treatments; (2) stress-corrosion cracking; (3) formation of a brittle sigma phase, particularly in the high-ferrite weld deposits which are frequently used to avoid weld cracking; (4) knife-line corrosion attack; (5) high thermal stresses due to poor heat conductivity; and (6) accelerated local attack by highly corrosive, rapidly flowing solutions due to removal of protective films.

8.189. The approximate compositions of a number of stainless steels which may find application in reactor operations involving solutions, e.g., for homogeneous reactors and chemical processing plants, are given in Table 8.15. Type

TABLE 8.15. STAINLESS STEELS FOR REACTOR APPLICATIONS

Type	Carbon, % (maximum)	Chromium, %	Nickel, %	Other Elements
304	0.08	18.0 to 20.0	8.0 to 11.0	—
304L	0.03	18.0 to 20.0	8.0 to 11.0	—
309S Cb	0.08	22.0 to 26.0	12.0 to 15.0	Cb (min. 8 × C)
316	0.10	16.0 to 18.0	10.0 to 14.0	Mo (2.0 to 3.0%)
316L	0.03	16.0 to 18.0	10.0 to 14.0	Mo (1.75 to 2.5%)
347	0.08	17.0 to 19.0	9.0 to 12.0	Cb (min. 10 × C)

304, which is a familiar variety, can be considered if the environment does not promote intergranular attack of sensitized stainless steel, if the structure is not to be welded, or if the welded structure can be given a full anneal at about 1000°C (1830°F), followed by fast cooling to avoid sensitization. On the other hand, where intergranular attack of the sensitized material is to be expected or if field welding precludes annealing, Types 304L (low carbon) and 347 may prove useful.

8.190. Nickel alloys, such as Nichrome, Monel metal, Inconel, and Hastelloys, are also of interest, especially since they are fairly resistant to attack by fused salts and alkali hydroxides. Copper-base alloys do not appear to be of value for reactor construction because of their high creep rate at elevated temperatures.

ZIRCONIUM AS A REACTOR MATERIAL*

Production of Zirconium

8.191. As far as can be seen at present, zirconium, alone or preferably in the form of alloys, seems to be a very promising metal for use as a constructional and fuel-cladding material in thermal reactors, at temperatures up to about 400°C (750°F). Until recently the scientific and technical literature contained relatively little information concerning this element, and so its production and some of its important properties will be described here. It may be mentioned that much of the development that has taken place in connection with zirconium and its alloys since 1947 has been a direct result of their potential use in reactor construction.

8.192. The chief zirconium mineral is the silicate *zircon*, $ZrSiO_4$, considerable deposits of which are found in Brazil, India, Australia, and Ceylon. It also occurs in appreciable quantities in various parts of the United States, including Florida, California, Oregon, and Idaho. Another commercial mineral is *baddeleyite*, an impure form of zirconium dioxide, ZrO_2, which is associated with zircon in the diamond-bearing sands of Brazil. Zirconium is by no means a rare element; it is the eleventh most common in the earth's crust, and is thus more plentiful than copper, lead, and zinc.

8.193. An important fact from the reactor standpoint is that zirconium ores invariably contain from 0.5 to 3.0 per cent of hafnium, and this element has a high cross section (115 barns) for the capture of thermal neutrons. It is desirable, therefore, that most of the hafnium be removed from zirconium for use in thermal reactors. Unfortunately, this is not a simple matter because the chemical properties of hafnium and zirconium are very similar; nevertheless, successful methods of separation have been developed.†

8.194. In the production of zirconium, the minerals are first separated from the accompanying gangue and other impurities by various combinations of gravity concentration, screening, and mechanical and electrostatic separation. The ore is then heated with carbon (graphite) in an electric furnace when zirconium carbide (or cyanonitride in the presence of atmospheric nitrogen) is formed. In this operation the silica is driven off. The carbide (or cyanonitride)

* For reviews of the production and properties of zirconium, see Symposium, "Zirconium and Zirconium Alloys," American Society for Metals, 1953; E. C. Miller, *Nucleonics*, **11**, No. 7, 27 (1953); *Metal Prog.*, **63**, No. 5, 67 (1953); A. D. Schwope, *ibid.*, **63**, No. 5, 75 (1953); H. A. Saller, *Proc. Conf. on Nuc. Eng.*, Berkeley, 1953, p. 11; W. A. Johnson, *Chem. Eng. News*, **32**, 1882 (1954); J. W. Holladay and J. G. Kura, *Chem. Eng. Prog. Symposium Series*, No. 11, **50**, 23 (1954); G. L. Miller, "Zirconium," Academic Press, Inc., 1954; C. A. Hampel (Ed.), "Rare Metals Handbook," Reinhold Publishing Corp., 1954, Chapter 28 (by A. W. Schlechten); "Metallurgy of Zirconium," National Nuclear Energy Series, Div. VII, Vol. 4, McGraw-Hill Book Co., Inc., to be published, 1955.

† Small proportions of hafnium remaining in the zirconium are not necessarily harmful, since they can serve as a consumable poison to provide partial compensation for the change in reactivity accompanying fuel consumption (see § 12.113).

is converted into zirconium tetrachloride, $ZrCl_4$, by means of chlorine gas, and the salt is then dissolved in water containing hydrochloric acid. After removal of the hafnium,* the zirconium is precipitated as a basic sulfate from a solution of pH 1.4 at 90°C (194°F). The basic zirconium sulfate is filtered off and digested with ammonium hydroxide to form zirconium hydroxide. (The latter can be obtained by direct addition of alkali or ammonium hydroxide to the zirconium

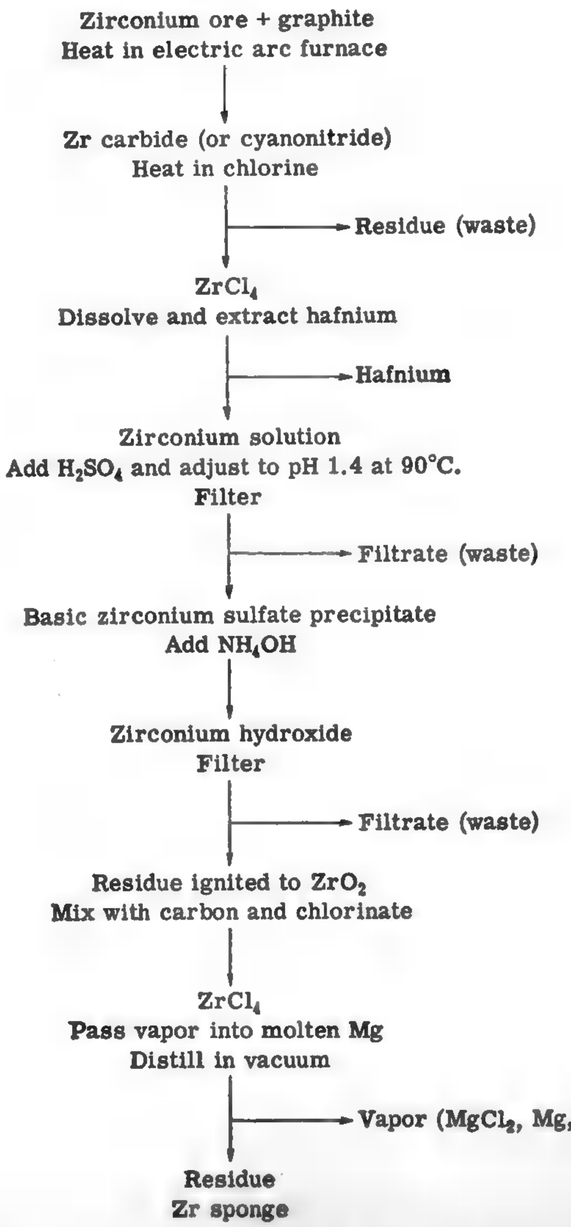


FIG. 8.15. Simplified flow sheet for extraction of zirconium from zircon

* The actual process used is classified, but various methods for separating hafnium from zirconium have been described in the unclassified literature, e.g., E. H. Huffman, *et al.*, Report UCRL-2536; see also W. M. Manning, Report AECD-3007, p. 9.

solution, but the resulting precipitate is difficult to filter.) The hydroxide precipitate is then filtered, washed, and ignited to form the oxide, ZrO_2. This is made into briquettes with carbon and chlorinated at high temperature (over 800°C or 1470°F); the zirconium tetrachloride formed, which sublimes at about 300°C (572°F), is collected in a condenser. The tetrachloride is then resublimed and the vapor passed over a bath of molten magnesium, when the reaction

$$ZrCl_4 + 2Mg \rightarrow Zr + 2MgCl_2$$

takes place. The magnesium chloride formed and excess magnesium are removed by vacuum distillation, leaving a mass of zirconium sponge.* A simplified flow sheet of the process for the production of zirconium is given in Fig. 8.15.

8.195. For use in reactors the zirconium sponge was at one time purified by the deBoer (or iodide) process, similar to that described in § 8.35 for thorium. Zirconium tetraiodide, formed by the interaction of iodine vapor with zirconium sponge, is decomposed on an electrically heated zirconium wire filament. The zirconium metal, in the form of pure "crystal bar," deposits on the filament, and the iodine is regenerated to react with more zirconium sponge, and so on. In the course of time a substantial bar of zirconium forms and can be removed for fabrication.

8.196. As result of improvements in the magnesium-reduction process, zirconium sponge of such high purity is obtainable that further treatment by the expensive iodide procedure is now regarded as unnecessary. There is still the problem of consolidating the sponge into massive metal, since molten zirconium attacks all known crucible materials. Graphite crucibles are moderately satisfactory, but enough carbon is introduced into the zirconium to affect its properties adversely. As a result of considerable research, an arc-melting process, using a consumable zirconium electrode and a water-cooled copper crucible to hold the melt, has been developed. The zirconium sponge, together with any desired alloying elements, is compacted into bars; these are welded together to form the consumable electrodes which are arc-melted to yield 6-in. ingots. To improve the homogeneity of the metal, the 6-in. ingots are joined by a threaded nipple and once more arc-melted to produce 10-in. ingots. By carrying out the arc-melting in an atmosphere of helium and argon the contamination by oxygen can be held down to 0.03 per cent and that by nitrogen to 0.001 per cent or less. These elements increase the tensile strength of zirconium but tend to make it brittle and therefore hard to fabricate. The presence of nitrogen is especially detrimental to corrosion resistance in water at high temperatures.

General, Physical, and Mechanical Properties of Zirconium

8.197. Pure zirconium is a silvery-white metal, its theoretical density being 6.49 g/cm³ at 20°C (68°F). Between room temperature and its melting point

* The product is sometimes called Bureau of Mines (or Kroll process) zirconium because it was developed at the Albany (Oregon) Laboratory of the Bureau of Mines, from a magnesium-reduction method initiated by W. J. Kroll.

(1845°C or 3355°F) it exists in two allotropic modifications: a close-packed hexagonal (alpha) phase, stable up to 863°C (1585°F), and a body-centered cubic (beta) phase stable above this temperature. In spite of its hexagonal (anisotropic) structure, the alpha form of zirconium can be prepared in a ductile form having mechanical and machining properties similar to those of plain carbon steel.

8.198. The physical properties of zirconium, e.g., thermal expansion and conductivity, vary with the nature and treatment of the metal. Anisotropic behavior is observed, as expected, both in single crystals and in rolled products. At ordinary temperatures the linear coefficient of thermal expansion is 10.3×10^{-6} per °C in the direction of the hexagonal axis and 4.5×10^{-6} per °C at right angles. The thermal conductivity decreases from 0.050 cal/(sec)(cm²)(°C/cm) at 25°C (78°F) to 0.045 at 300°C (572°F).

8.199. At temperatures up to about 400°C (750°F) relatively pure (e.g., crystal bar) zirconium and certain of its alloys, notably those containing small amounts of tin, are highly resistant to corrosion by air and water (or steam); a thin, stable, adherent oxide film is formed on the metal and this protects it from further attack. Above 400°C (750°F) it reacts more readily with oxygen and water and even with nitrogen, and since it loses a large part of its mechanical strength as well at these high temperatures, the use of zirconium as a structural material will probably be limited to lower temperatures. However, because of its excellent corrosion resistance and good mechanical properties at the required operating temperatures, in addition to its small cross section for neutron capture, zirconium was selected as one of the components of the core of the Submarine Thermal Reactor (Mark I).

8.200. Zirconium forms alloys with many other metals, and some of these are stronger and have greater resistance to corrosion than pure (crystal bar) zirconium. It is probable, therefore, that zirconium alloys will play an important part as structural and cladding materials for nuclear reactors in the future.

8.201. The mechanical properties of different specimens of zirconium vary widely because they are appreciably affected by treatment of the metal and by

TABLE 8.16. MECHANICAL PROPERTIES OF ZIRCONIUM

Property	Temperature		
	20°C (68°F)	210°C (410°F)	320°C (608°F)
Tensile strength, 10^3 psi	32	20	16
Yield strength (0.2% offset), 10^3 psi	18	10	8
Elongation, %	25	50	60
Modulus of elasticity, 10^6 psi	14	12	11
Poisson's ratio	0.35		
Hardness, Rockwell A	30		

the presence of traces of impurities. Although the data in Table 8.16 cannot be regarded as definitive, by any means, they provide some general indication of the mechanical properties of the relatively pure metal and their variation with temperature. It is seen that both the tensile strength and the yield strength fall off as the temperature is increased; the creep strength also decreases at the same time. These facts account for the statement made above that the use of zirconium as a structural material may be limited to temperatures below about 400°C (750°F). However, definite improvement can be achieved by alloying with other elements, such as aluminum, tin, molybdenum, and columbium (niobium).

8.202. It is noteworthy that small amounts of oxygen and nitrogen produce marked effects upon the mechanical properties of zirconium metal. For example, 0.1 per cent by weight of oxygen increases the tensile strength to about 42,000 psi, the yield strength for 0.2 per cent offset to 26,000 psi, and the hardness to Rockwell A 38; however, the elongation is decreased at the same time to about 14 per cent, so that the metal is less ductile. The hardening effect of oxygen and nitrogen (and of carbon) decreases as the temperature is raised and becomes negligible around 400°C (750°F), so that these elements do not improve the high-temperature properties of zirconium.

8.203. Zirconium metal is very susceptible to hydrogen embrittlement; as little as 10 ppm can produce a noticeable decrease in the Charpy V-notch impact value at normal temperatures. At higher temperatures, however, around 300°C (480°F), the effect becomes negligible. Hydrogen may be absorbed by zirconium both during production and in subsequent processing operations, but it can be removed by annealing in a good vacuum at 800 to 900°C (1470 to 1650°F).

8.204. All conventional fabrication methods, such as machining, hot- and cold-rolling, forging, extrusion, and drawing, can be used with zirconium, provided reasonable precautions are taken to prevent oxidation. For shapes which are to be machined to final size even this is not necessary. Large ingots can be subjected to hot-forging, blooming, and rolling in commercial steel-mill equipment without protection from the air. The relatively small surface contamination can be removed by the sand-blasting and pickling procedures in general use for the treatment of stainless steel. On the other hand, pieces which are to be fabricated to close tolerances should be protected by an inert atmosphere or by jacketing with copper or steel. Such jacketing is also necessary for the extrusion of zirconium because the metal tends to gall in the die. Alternatively, glass can be used both to protect and lubricate zirconium during extrusion.

8.205. Because hot operations have the advantage of easy size reduction, zirconium ingots are generally forged and then hot-rolled as the initial operation. After cleaning the surface, the metal can be readily cold-worked, followed by annealing in vacuum or in an inert atmosphere to prevent attack and contamination by oxygen and nitrogen from the air. After annealing, cold-worked zir-

conium is very ductile (provided oxygen, nitrogen, and carbon have been kept out) and can be machined readily.

8.206. Powder-metallurgical methods can also be used to fabricate zirconium parts.* Since zirconium itself is too ductile at ordinary temperatures for comminution by mechanical methods, it is first converted into the hydride, e.g., by the action of hydrogen gas on zirconium sponge or crystal bar at 400 to 800°C (750 to 1470°F). The hydride is a brittle material which can be readily pulverized in hammer or pebble mills, and the resulting powder can be decomposed into zirconium powder and hydrogen upon heating in vacuum. For fabrication purposes the metal powder obtained in this manner may be compacted at pressures of about 50 tons/in.^{2} and then sintered at temperatures over 1000°C (1830°F). In order to prevent access of oxygen and nitrogen the sintering process is carried out either in the presence of a pure inert gas or under vacuum. Zirconium hydride powder itself may be compacted directly and then sintered in a vacuum furnace; decomposition of the hydride takes place during sintering and the resulting product is superior in many ways to that obtained from zirconium powder. Samples of zirconium made from powder as just described have highly satisfactory mechanical properties. They are hard and ductile and can be cold-rolled without difficulty.

8.207. Zirconium metal has excellent welding characteristics, but the operation must be performed in an atmosphere of inert gas of high purity, e.g., helium or argon, to prevent contamination. The ability of molten zirconium to dissolve surface films of oxide and nitride is an important factor in the production of bonds having a high degree of integrity. Soldering and brazing by conventional methods are satisfactory, provided certain precautions are taken.

Corrosion Resistance of Zirconium

8.208. One of the outstanding properties of zirconium is its resistance to corrosion by the great majority of acids, alkalies, and salts in aqueous solution under all reasonable conditions. Among the substances which attack the metal are hydrofluoric acid, concentrated sulfuric acid, phosphoric acid, and ferric and cupric chloride solutions. From the reactor standpoint the resistance to corrosion and erosion in water at high temperatures is important; a 500-hour dynamic test in water at 205°C (401°F) indicated that zirconium suffered less attack than stainless steel, columbium (niobium), beryllium, carbon steel, and aluminum, the superiority over the other metals increasing in the order given. It should be pointed out, however, that the corrosion-erosion behavior of zirconium is markedly dependent on the composition of the material, as will be seen below, on its metallurgical history, and on the state of its surface, as well as on the purity of the water and the presence of dissolved oxygen and nitrogen.

* H. H. Hausner, H. S. Kalish, and R. P. Angier, *Trans. Am. Inst. Mining Met. Engrs.*, **191**, 625 (1951); H. H. Hausner and H. B. Michaelson, *Chem. Eng. Prog. Symposium Series*, No. 11, **50**, 11 (1954).

8.209. The corrosion behavior of zirconium in water at high temperatures is very sensitive to the presence of impurities.* Of the elements which are likely to be present, nitrogen, carbon, aluminum, and titanium are particularly deleterious. Other harmful elements are oxygen, magnesium, silicon, and calcium, but these are generally found in zirconium in amounts too small to be detrimental. In small concentrations, up to about 0.1 per cent, copper, tungsten, iron, chromium, and nickel appear to have little influence on the corrosion of zirconium by high-temperature water.

8.210. Because the production of zirconium metal of a very high degree of purity is expensive, attempts have been made to improve the corrosion resistance of the commercial, e.g. arc-melted, product by the addition of various alloying metals. In this connection alloys containing 1.5 per cent or less of tin show considerable promise, especially if small quantities of iron, nickel, or chromium are present in addition. These alloys also have good mechanical properties at high temperatures.

8.211. The resistance of zirconium to attack by certain liquid metals is of importance because of their possible use as reactor coolants (§ 8.242). Sodium and sodium-potassium alloys have little effect up to temperatures of 600°C (1110°F), provided the oxygen content is low. The resistance to lithium, bismuth-lead alloy, and bismuth-lead-tin alloy is reported to be good at 300°C (570°F) but limited (or moderate) at 600°C (1110°F).†

CERAMIC MATERIALS FOR REACTORS‡

General Characteristics of Ceramics

8.212. The term "ceramics" is used in a very general sense to refer to materials of various types, ranging from graphite to cement, obtained by processes involving the use of high temperatures. Because of their great thermal stability and refractory nature, ceramic materials appear to be especially suited for use in reactors operating at elevated temperatures. Such reactors are of particular interest for power production because of the increase in thermal efficiency which is possible from their use.

8.213. In comparison with metals, ceramics have the advantages of ability to withstand higher operating temperatures and better resistance to corrosion by various gases and liquids which might be used as reactor coolants. On the other hand, the thermal conductivity and tensile strength of ceramic materials are usually less than for metals. The discrepancy between the strengths of metals, on the one hand, and of ceramics, on the other hand, becomes less marked

* D. E. Thomas, *Chem. Eng. Prog. Symposium Series*, No. 12, **50**, 16 (1954); Report WAPD-T-186.

† R. N. Lyon (Ed.), "Liquid-Metals Handbook," U. S. Government Printing Office, 1952, Chapter 4 (by E. C. Miller).

‡ R. F. Geller, *Nucleonics*, **7**, No. 4, 3 (1950); F. H. Norton, Report AECD-3424; J. F. White, *Ceram. Soc. Bull.*, **32**, 301 (1953); see also, *Nucleonics*, **11**, No. 7, 20 (1953).

as the temperature is raised. It has been reported, for example, that few materials are as strong as graphite above 1700°C (3090°F). Ceramic materials are, in general, brittle at ordinary temperatures, and this brittleness cannot be overcome, as it can be frequently with metals, by proper treatment. Consequently they present a difficult fabrication problem. However, at higher temperatures ceramics may develop plastic or elastico-viscous properties, so that they become much less brittle.

8.214. One of the aspects of ceramics about which little is as yet known is the effect upon them of nuclear radiations, especially neutrons and fission products, at high temperatures. It is generally accepted that radiation damage will be greater than in metals, since most ceramics are chemical compounds with no free electrons. In ceramics of the ionic type, radiation will probably cause atomic (or ionic) displacements only, and so there should be self-healing or annealing, as is the case with metals, at elevated temperatures. But if covalent bonds are broken by the radiation, it is possible that the damage may be increased when the temperature is raised. Further, if there are any atomic-migration reactions which might occur in the ceramic material, then it is to be expected that, as with metals (§ 8.109), they will take place at a lower temperature in the presence of nuclear radiation.

8.215. In view of the interest in high-temperature operations, e.g., for jet engines as well as for nuclear reactors, studies are being made of combinations of ceramic materials and metals; these mixtures have been called "cermets." It is hoped to combine in them the thermal stability and refractory character of ceramics with the strength, ductility, and thermal conductivity of metals. Among the cermets which have attracted attention mention may be made of the systems Al_2O_3-Al, Al_2O_3-Cr, ZrC-Fe, TiC-Ni, and SiC-Si. In the latter the element silicon, which is not a metal, takes the place of a metal.

Applications of Ceramic Materials

8.216. Ceramics have actual or potential use in nuclear reactors as fuel elements, moderators, shielding, control devices, structural materials, and insulators. Graphite, beryllium oxide, cement (in concrete), and uranium dioxide have already been employed, or considered for employment, in reactors, either existing or planned. There are, however, a number of other ceramics or cermets which might well find application (or be given serious thought) in the future. Shielding materials will be discussed in Chapter X, and moderators have already been given some consideration. The treatment here will, therefore, be restricted mainly to other uses of ceramic materials.

Fuel Elements

8.217. In addition to the fact that it is readily attacked by air and water, uranium suffers from the disadvantages of undergoing phase changes at 662 and 772°C (1224 and 1422°F) and of having only a moderately high melting point

(1133°C or 2070°F). It is apparent, therefore, that, for high-temperature operation, reactor fuel elements of metallic uranium will be unsatisfactory. Various uranium compounds, such as the dioxide (UO_2), the nitride, carbides, and silicides, which are ceramic-like in character, have been proposed as alternatives, since they are stable up to temperatures of about 2000°C (3630°F) or more. Combinations of these substances with more conventional ceramic materials, including magnesium and beryllium oxides and silicon carbide, have also been suggested for the production of fuel elements for use at elevated temperatures.

8.218. In addition to temperature stability, it is necessary that a fuel element maintain reasonable strength over a considerable temperature range. Further, it should have good corrosion resistance, a reasonable capacity to retain fission products, and the ability to withstand the action of nuclear radiations. Finally, the substances present, apart from uranium, should have relatively low absorption cross sections for neutrons. Although these requirements limit the choice of ceramic materials for use in reactor fuel elements, there are a few which satisfy some of them, at least, as noted above.

Structural Materials

8.219. Some properties of a number of ceramics and cermets which might have some application as structural materials for reactors operating at high temperatures are given in Table 8.17. Most of the data apply to normal temperatures, since these are the only ones at present available. In any event, the physical and mechanical properties are extremely variable; they are dependent upon the method of fabrication of the specimen under examination, the amount and nature of minor additions, and the firing temperature.

TABLE 8.17. PROPERTIES OF CERAMIC MATERIALS FOR REACTOR APPLICATION

Material	Melting Point (°C)	Melting Point (°F)	Theoretical Density (g/cm³)	Thermal Conductivity [cal/(sec)(cm²)(°C/cm)]	Thermal Expansion Coefficient (per° C × 10⁶)	Tensile Strength (10³ psi)	Modulus of Elasticity (10⁶ psi)
Al_2O_3	2030	3690	3.96	0.04	7	36	52
BeO	2520	4570	3.0	0.5	8	14	45
MgO	2800	5070	3.6	0.05	13	~2	12
ZrO_2	2700	4890	6.3	0.004	6	18	36
SiC	2200*	3990	3.2	0.08	4	~2	—
SiC-Si	1500*	2730*	2.5	0.4	3	10	—
Al_2O_3-Cr	1400**	2550**	5.9	0.04	6	39	47
TiC-Ni	1400**	2550**	5.8	0.08	4	90	—

* Decomposition temperature.
** Maximum service temperature.

8.220. Two substances appear to be of special interest for use in reactors at high temperatures; these are graphite, whose properties were recorded in Table 8.12, and beryllium oxide. In addition to being good moderators, with small absorption cross sections for neutrons, they have high thermal conductivities and excellent resistance to thermal shock. Although the tensile strength of graphite is small, it does increase with temperature; on the other hand, that of beryllium oxide decreases with increasing temperature. An important advantage of the latter substance is its greater resistance to corrosion or attack by other substances which might be required in reactor operation.

Other Uses of Ceramic Materials

8.221. The materials chiefly used for the control of thermal reactors at moderate temperatures are cadmium, boron steel, and, to some extent, boral (B_4C-aluminum). None of these is, however, suitable for high-temperature applications. Compounds of boron which are stable at elevated temperatures are boron carbide (B_4C) and boron nitride (BN), and both could be employed for reactor control. Among other ceramic materials having large cross sections for the absorption of thermal neutrons are hafnium dioxide (HfO_2), which is obtained in connection with the extraction and purification of zirconium (§ 8.194). Since a number of the rare-earth elements are excellent neutron absorbers, thermally stable control devices might be prepared by the addition of the oxides to suitable ceramic materials.

8.222. Finally, mention may be made of the use of ceramics as heat insulators, e.g., for the pipes carrying hot coolant from the reactor to the heat exchanger. However, such materials do not need to have properties that differ in any way from conventional insulating materials except, perhaps, the ability to withstand higher temperatures.

REACTOR COOLANTS

General Characteristics*

8.223. Since most of the energy liberated in fission appears as heat, a nuclear reactor must have an adequate cooling system which will prevent the attainment of undesirable temperatures within the reactor. Such temperatures may be determined by a property of the fuel, e.g., uranium metal undergoes a phase change at 662°C (1224°F) accompanied by a relatively large increase in volume (§ 8.37), or of the coolant, e.g., it may be undesirable in a water-cooled reactor for the water to boil. The cooling medium (or coolant) may be gaseous or liquid, and the specific requirements for this substance will depend largely upon the rate of heat production, i.e., the reactor power density, and the operating

* O. J. Woodruff, Jr., J. W. McShane, and W. J. Purcell, *Nucleonics*, **11**, No. 6, 27 (1953); E. Luebke, *Proc. Conf. on Nuc. Eng.*, Berkeley, 1953, F-41; W. E. Parkins, *ibid.*, F-55; T. Shimazaki, *Chem. Eng. Prog. Symposium Series*, No. 12, **50**, 113 (1954).

temperatures. For a reactor of low power output, in which no use is made of the heat generated, the cooling system may be relatively simple. On the other hand, for economic power production a reactor should operate at high temperatures, and then careful consideration must be given to the choice of coolant.

8.224. In general a coolant should possess the following characteristics: (1) good thermal properties, e.g., high specific heat and thermal conductivity; (2) low power requirement for pumping; (3) high boiling point and low melting point; (4) stability to heat and radiation; (5) suitable corrosion characteristics in the given system; (6) small cross section for neutron capture; (7) nonhazardous, including low induced radioactivity; and (8) low cost. These topics will be reviewed briefly in the order given, although this must not be regarded as representing the order of their importance. In actual practice the coolant chosen for any particular application will represent a compromise among conflicting requirements.

8.225. Since the primary purpose of a coolant is the removal of heat from a reactor, it must possess good heat-transfer properties. For a given geometry and dynamics of flow this condition is best met by high thermal conductivity and high specific heat; for nonmetallic liquids in particular, low viscosity is also advantageous in this connection. The fraction of the power output required to pump the coolant through the reactor and heat exchanger should be small, and this requires a coolant with a high density and low viscosity. For use at high temperatures a liquid coolant should have a low vapor pressure, so that a strong pressurized system is not necessary. At the same time, the coolant should remain liquid down to low temperatures to avoid the possibility of solidification when the reactor is shut down. These two requirements are, to a certain extent, contradictory; the former implies a high boiling point and the latter a low melting point. Nevertheless, a number of substances are known, e.g., liquid metals and molten salts, that have a very wide temperature range for the liquid phase. The problem considered here does not arise, of course, when a gas is used as coolant.

8.226. A coolant should be stable with respect both to high temperatures and to the action of nuclear radiation. If the reactor has a circulating fuel, so that the secondary coolant is not exposed to a very high radiation field, the problem of radiation decomposition (and of induced radioactivity) is not so serious as when the coolant actually flows through the reactor. Further, it is necessary that the fluid should not attack the materials, either inside or outside the reactor, with which it comes into contact. Since the coolant system is a dynamic one, mass transfer due to temperature gradients may be very significant. As far as physical and chemical attack is concerned, the coolant and containing materials must, of course, be considered together.

8.227. Because loss of neutrons must be compensated for by an increase in the fuel loading of the reactor, the coolant should consist of an element or elements having small cross sections for neutron capture. This restriction is not im-

TABLE 8.18. PHYSICAL PROPERTIES OF COOLANT MATERIALS

Coolants	M.P., °C	B.P., °C	Temp., °C	Density, g/cm³	Sp. Ht., cal/g	Thermal Cond., cal/(sec)(cm²)-(°C/cm)	σ_a, barns per nucleus	Induced Activity
Hydrogen (1 atm)	—	Gas	100 300	6.6×10^{-5} 4.3×10^{-5}	3.43 3.50	5.33×10^{-4} 7.36×10^{-4}	0.33	None
Helium (1 atm)	—	Gas	0 100	1.8×10^{-4} 1.4×10^{-4}	1.25 1.25	3.3×10^{-4} 4.0×10^{-4}	~0	None
Air (1 atm)	—	Gas	100 300	9.5×10^{-4} 6.2×10^{-4}	0.240 0.242	7.57×10^{-5} 1.09×10^{-4}	1.5	N^{16} 7.3s; 6-Mev γ A^{41} 1.8h; 1.4-Mev γ
Carbon dioxide (1 atm)	—	Gas	100 300	1.5×10^{-3} 9.5×10^{-4}	0.218 0.231	5.0×10^{-5} 9.0×10^{-5}	0.003	N^{16} (as above)
Water (liquid)	0	100 (1 atm) 250 (575 psia)	100 250	0.958 0.794	1.01 1.10	1.70×10^{-3} 1.96×10^{-3}	0.22	N^{16} (as above)
Lithium	179	1317	200 600	0.507 0.474	1.0 1.0	0.09	0.033 for Li^7	Li^8 0.83s; bremsstrahlung from 13-Mev β
Sodium	98	883	100 400	0.928 0.854	0.331 0.306	0.206 0.170	0.50	Na^{24} 15h; 1.38-, 2.75-Mev γ
Sodium (22 wt %)-potassium (78 wt %) alloy	−11	784	100 400	0.775 0.703	0.223 0.210	0.0583 0.0636	1.7	Na^{24} (as above) K^{42} 12.4h; 1.5-Mev γ
Lead (44.5 wt %)-bismuth (55.5 wt %) eutectic	125	1670	200 600	10.46 9.91	0.035 ~0.035	0.023 ~0.035	0.1	Bi^{210} 5d; decays to toxic Po^{210}
Lead (97.5 wt %)-magnesium (2.5 wt %) eutectic	250	~1700	300	~10	~0.035	~0.03	0.16	Mg^{27} 10m; 0.84-, 1.0-Mev γ
Sodium hydroxide	323	—	350 550	1.79 1.67	0.554 0.434	5.2×10^{-3} 2.8×10^{-3}	0.28	Na^{24} (as above) N^{16} (as above)

portant for fast reactors, because the capture cross sections for high-energy neutrons are invariably small. If, as a result of neutron capture, the (primary) coolant acquires induced activity, this should be weak with little or no gamma radiation in order to avoid the necessity for shielding pumps, heat exchanger, and piping. It is desirable, in any event, that the cooling medium not be toxic or otherwise hazardous, and that it does not become a hazard as a result of exposure to radiation. Finally, the coolant should be readily available at relatively low cost.

8.228. For a thermal reactor it is desirable, too, that the elements present in the coolant should have low atomic weights. In other words, it is an advantage if the coolant acts also as a moderator and makes some contribution to the slowing down of the neutrons. On the other hand, for a fast reactor, where slowing down is to be avoided, materials of low mass number should be eliminated, as far as possible.

8.229. It will be seen shortly that no single substance (or mixture) satisfies all the requirements laid down for a satisfactory reactor coolant. Before considering various individual coolants, the most important properties of those substances which appear to offer some prospect for use in reactors, now or in the future, are summarized in Table 8.18.* Because a coolant may be used over a range of temperatures, values are given for two temperatures where possible.

Gaseous Coolants†

8.230. On the basis of general radiation and thermal stability, ease of handling, and absence of hazardous conditions, there is much to recommend the use of a gas as coolant. Since it is readily available, air is the most obvious choice. Unfortunately, air is not a good heat-transfer material and, as with gases in general, a large proportion of the power produced is consumed in pumping the air through the cooling system. Further, at high temperatures the oxygen (and in some cases the nitrogen also) is likely to attack the graphite or beryllium moderator and the structural and fuel-canning materials. Air is used as the coolant in several of the natural uranium-graphite reactors (see Table 1.10), but these are all large and operate at relatively low power densities. The heat flux is thus fairly small and excessive volumes of air are not needed to keep the fuel temperatures at reasonable levels.‡ For research reactors such as these, the inefficiency of air as a coolant is offset by the simplicity of design. Air is also used as coolant in the British plutonium-production reactors;§ this choice was determined largely by safety considerations and also because of the difficulty

* More complete data, expressed in standard engineering units, will be found in the Appendix.

† See J. B. Haddow, Report on Atomic Power Symposium, Chalk River, Ontario, 1953 (CRR-548-A), p. 110.

‡ The maximum heat flux, near the center, of the Oak Ridge graphite reactor is 9250 Btu/(hr)(ft²) and the power density in the fuel is then about 120 kw/ft³.

§ K. E. B. Jay, "Britain's Atomic Factories," H. M. Stationery Office, 1954, p. 22.

in finding a large supply of reasonably pure water for cooling purposes, such as is available at Hanford. After leaving the reactor, the air is always somewhat radioactive, mainly due to the presence of argon-41, but since it is unnecessary to use a closed system, the air is discharged through a high stack.

8.231. Although hydrogen would be the best gaseous coolant, as far as both heat transfer and pumping power are concerned, it would constitute a serious explosion hazard if it escaped into the air. In addition its containment, especially under pressure, would require the use of special steels not subject to attack or embrittlement by hydrogen. Consequently no proposal has been made for the use of hydrogen as a reactor coolant.

8.232. From several points of view, helium has much to recommend it as a gaseous coolant. Although its thermal properties are not quite as good as those of hydrogen, it has a negligible cross section for neutron capture and is not hazardous. It is also stable to heat and to the action of radiation. In addition it is inert chemically and has no harmful effects on structural or containing materials. Because of the high cost of helium, it would be necessary to use a closed circulating system, and in order to economize in pumping power moderately high (or high) pressures would be necessary, thus introducing the problem of leak-proof pressurized vessels. It is of interest to mention that, when the Hanford reactors were first designed, helium was the preferred coolant; it was not used, however, mainly because of the difficulties anticipated in the prevention of leakage of helium gas under pressure from such large structures and in the loading and unloading of the reactor fuel.* More recently a proposal has been made to use helium at 10 atm pressure as the coolant in a dual-purpose reactor for production of both plutonium and power.†

8.233. Nitrogen gas at 10 atm pressure was originally the coolant in the French (P2) reactor at Saclay, but this is being abandoned in favor of carbon dioxide. Not only has nitrogen a fairly large neutron-capture cross section, but as a result of the (n, p) reaction radioactive carbon-14 is formed which would constitute a hazard if it escaped from the pressure vessel. Safety considerations have also determined the use of carbon dioxide under pressure as the coolant for the British central-station power reactor, which is expected to begin operation in 1956 or 1957.‡ The carbon and oxygen nuclei in carbon dioxide have small cross sections for neutron capture, and the gas itself is free from the dangers of toxicity or explosion. Carbon dioxide does not readily attack metals, although it does react with graphite at fairly high temperatures. It is less efficient than helium as regards heat transfer, pumping power, and neutron absorption, but it is, of course, much more readily available. An uncertain factor in connection with the use of carbon dioxide as a coolant is the effect of

* H. D. Smyth, "Atomic Energy for Military Purposes," U. S. Government Printing Office, 1945, § 7.16.

† "Reports to the U. S. Atomic Energy Commission on Nuclear Power Reactor Technology," U. S. Government Printing Office, 1953, p. 1; see also *Nucleonics*, **11**, No. 6, 51 (1953).

‡ See *Atomics*, **5**, 174, 275 (1954).

the intense radiation field in the reactor upon its stability and interaction with graphite and other materials.

Water as Coolant

8.234. From the standpoint of cost, water ranks next to air for use as a coolant, provided it is available in a sufficient degree of purity. Thus one of the controlling factors in the location of the Hanford and Savannah River plants was the accessibility of large volumes of water of unusual clarity and purity. Nevertheless, some further purification is necessary, as will be seen below, which adds to the cost. Water has a high specific heat, although only a fair thermal conductivity as compared to liquid metals; it has a relatively low viscosity and is easily pumped. The pumping power for equivalent heat removal is roughly on the order of one tenth of that required with a gaseous coolant at 10 atm pressure. One of the great advantages of water, of course, is that it can act simultaneously as moderator and coolant, e.g., in the MTR, STR, and PWR.

8.235. The use of water as coolant has, however, a number of drawbacks. Among these may be noted a fairly large neutron-capture cross section, decomposition by radiation, corrosive action on metals, and low boiling point at normal pressures. Further, the induced radioactivity, especially that of nitrogen-16, resulting from the (n, p) reaction of fast neutrons with oxygen-16, as well as that due to the capture of thermal neutrons by impurities, e.g., sodium, must not be overlooked. The loss of neutrons can be compensated by loading additional fuel into the reactor in order to attain criticality or by the use of uranium somewhat enriched in the 235-isotope, e.g., in the MTR, PWR, etc. The effect of radiation on water is fairly well understood and the extent of decomposition can be minimized by removing ionic impurities, a procedure which is desirable for other reasons. If decomposition is appreciable, a degasifier may be necessary to remove the oxygen and hydrogen gases formed.

8.236. At moderate temperatures enough data concerning the corrosive properties of water are available to permit the design of satisfactory cooling systems. Aluminum can be used as the piping material if the temperatures are moderate and stainless steels are available for higher temperatures. In order to minimize corrosion and the formation of scale on the heat-transfer surfaces, the water must be purified, by demineralization or distillation. In some circumstances deaeration may also be necessary. The Columbia River water used to cool the Hanford reactors is purified for an additional reason—to decrease the amount of radioactivity introduced into the river by the return water.

8.237. For the efficient production of useful power, a reactor must be operated at temperatures well above the normal boiling point of water under atmospheric pressure. In these circumstances, as in the STR and PWR, for example, high pressures must be used, which means the reactor must be enclosed in a pressure vessel. Further, at high temperatures even pure water, free from ionic impurities and oxygen, becomes very corrosive. As a result of the corrosion

problem and the need for high pressures, the use of water as coolant in a power reactor imposes limitations upon materials and equipment. Among the materials which possess the necessary properties of strength and corrosion resistance are certain stainless steels, titanium (or its alloys), Inconel, and K Monel metal. In addition, zirconium has a number of useful applications.

8.238. Although it has been realized for some time that boiling water (or other liquid) would be effective as a coolant, because advantage would be taken of the latent heat of vaporization, it was felt that bubble formation in a reactor would lead to instability that might result in a catastrophic increase of reactivity. However, it has now been shown that it is not at all difficult to design a reactor, with solid fuel elements and water as coolant and moderator, in which bubble formation causes a decrease, rather than an increase, of reactivity. This effect is due largely to the decrease in the resonance escape probability (if moderately enriched fuel is used) and to the increase in neutron leakage accompanying the partial expulsion of the moderator.

8.239. Because of the self-regulating effect of steam-bubble formation, a reactor can be operated continuously in a stable fashion under boiling conditions. If the water boils more rapidly than required by the normal operating power, the additional bubble formation will decrease the reactivity and less heat will be produced by fission. This will tend to bring the boiling rate back to the desired value. The great advantage of a boiling-water reactor of this type is that it would produce steam directly, without the use of a heat exchanger, so that the same over-all efficiency for power production could be attained with lower temperatures and pressures in the reactor and greatly reduced pumping power.*

Heavy Water as Coolant

8.240. The only significant difference between ordinary water and heavy water as a reactor coolant (or coolant-moderator) lies in the appreciably smaller cross section of the latter for the capture of thermal neutrons. The improvement in neutron economy means that less fissionable material is required to make the reactor critical and natural uranium can be used as the fuel. In a few research reactors, including CP-3′ and CP-5 at Argonne National Laboratory, the heavy-water moderator is circulated through a heat exchanger so that it is also the coolant. Such reactors operate at relatively low power densities, and the total volume of heavy water is not excessively large. A proposal has been made to utilize this substance as the coolant-moderator in a dual-purpose reactor capable of a (gross) electric power output of 225,500 kw, but the high cost of the heavy water makes the design impractical at the present time.†

* "Report on the Five-Year Power Reactor Development Program Proposed by the Atomic Energy Commission," U. S. Government Printing Office, 1954, pp. 11, 15; S. Untermyer, *Nucleonics*, **12**, No. 7, 43 (1954); W. H. Zinn, Atomic Industrial Forum Report, July 1954, p. 36.

† "Reports to the U. S. Atomic Energy Commission on Nuclear Power Reactor Technology," U. S. Government Printing Office, 1953, p. 17. The cost of the heavy water alone, based on the market price of $80 per lb, would amount to about $180 per kw of electrical output for the particular design described; this is about as much as the total cost of a conventional coal-burning plant.

8.241. If heavy water should become available at a reasonable price, as it may well do in due course, a particularly interesting application would be as coolant-moderator in a boiling-water type of reactor. It is probable that such a reactor would be self-regulating under all conditions, irrespective of whether the fuel were natural or enriched uranium and of the ratio of uranium-235 to heavy water. Further, the greater spacing permissible between the fuel elements would facilitate the escape of steam in a boiling heavy-water reactor, as compared with one using ordinary water.

Liquid Metals as Coolants*

8.242. For reactors with high thermal fluxes, operating at high temperatures, liquid metals appear to have special interest as coolants. They have excellent thermal properties, e.g., high thermal conductivity and low vapor pressure; metals of low atomic weight, e.g., lithium and sodium, also have relatively high specific heats and volumetric heat capacities. Further, liquid metals are stable at high temperatures and in intense radiation fields. Their main disadvantage lies in difficulty of handling and the corrosive properties of some metals at high temperatures. It should be noted, however, that the General Electric Company has for several years used mercury in boilers, and that the Argonne National Laboratory and Knolls Atomic Power Laboratory have gained considerable experience with flow systems of such potentially hazardous coolants as liquid sodium-potassium alloy and liquid sodium, respectively. In any event, the corrosive properties of liquid metals do not become pronounced until temperatures at which other liquids are no longer usable as coolants.

8.243. The heat-transfer characteristics of several liquid metals are superior to those of water, so that it is possible to achieve the same rate of heat removal with smaller areas of contact between fuel and coolant. On the other hand, the volumetric heat capacities of liquid metals are less than that for water, so that a higher flow rate may be necessary to remove the same amount of heat. The pumping power depends on several factors, and it may be greater or less than for water; for the metals of low mass number it is generally somewhat less, but it is larger for elements of high mass number, e.g., bismuth and lead. On the whole, pumping power would not be an important consideration in the choice of a coolant among various liquid metals and water.

8.244. Some indication of the metals which might be used as coolants in thermal reactors can be obtained by selecting elements having relatively low absorption cross sections for thermal neutrons and melting points below 500°C (930°F). These are given in Table 8.19; as before, they are divided into two classes, namely, those with absorption cross sections less than 1 barn, and those with cross sections from 1 to 10 barns. Mercury is not included in this table because of its large cross section (340 barns) for the capture of thermal neutrons.

* R. N. Lyon (Ed.), "Liquid-Metals Handbook," U. S. Government Printing Office, 1952; T. Trocki, *Nucleonics*, **10**, No. 1, 28 (1952); H. E. Tibbs, Report on Atomic Power Symposium, Chalk River, Ontario, 1953 (CRR-548-A), p. 128; W. E. Parkins, *loc. cit.*

For fast neutrons the capture cross section is much less, and mercury was used as coolant in the Los Alamos Fast Reactor. However, the boiling point of mercury is 357°C (675°F), and its vapor pressure at moderate temperatures is relatively so high that it is considered less desirable than other possible coolants.

TABLE 8.19. METALS OF SMALL THERMAL-NEUTRON CROSS SECTION AND LOW MELTING POINT

Small Thermal Cross Sections				Intermediate Thermal Cross Sections			
		M.P.				M.P.	
Metal	σ_a, barns	(°C)	(°F)	Metal	σ_a, barns	(°C)	(°F)
Bismuth	0.032	271	520	Potassium	2.0	62	145
Lead	0.17	327	621	Gallium	2.7	30	86
Sodium	0.50	98	208	Thallium	3.3	302	576
Tin	0.65	232	450				

8.245. Of the elements in Table 8.19, potassium, gallium, and thallium do not present any notable advantages, especially as the two latter are expensive. Further, liquid gallium is difficult to contain at high temperatures, and the same is true of tin. Bismuth and lead have satisfactorily low cross sections, but their melting points are fairly high. A eutectic alloy of these two elements, containing 44.5 per cent of lead by weight, melts at 125°C (257°F) and may be a possible reactor coolant; another possibility is a lead-magnesium eutectic, containing 97.5 per cent of lead, which melts at 250°C (482°F).

8.246. A significant drawback to the use of bismuth in a reactor is the fact that it undergoes (n, γ) reaction to form bismuth-210 (radium-E), and this is a beta-particle emitter of 5.0 days half life, decaying to yield polonium-210. The latter constitutes an exceptionally insidious hazard because it is very toxic and is difficult to contain. Nevertheless, the problem is not regarded as insoluble, and a reactor has been designed with a circulating fuel of uranium dissolved in molten bismuth.* The best containing materials for liquid bismuth, and also probably for lead and its alloys, are chrome steels, e.g., $2\frac{1}{4}\%$ Cr-1% Mo and 5% Cr-$1\frac{1}{2}\%$ Si. Mass transfer, which becomes important at high temperatures, around 550°C (1020°F), can be controlled by addition of small amounts of zirconium and magnesium to the liquid metal.

8.247. Liquid sodium has attracted much attention as a reactor coolant, as will be seen shortly, as also has an alloy of 22 per cent (by weight) of sodium and 78 per cent of potassium, written NaK and generally referred to as "nack," which is liquid at room temperatures. Lithium is included in Table 8.18 in spite of the large capture cross section of the natural form of the element for

* See reports by members of the staff of Brookhaven National Laboratory, *Chem. Eng. Prog. Symposium Series*, No. 11, **50**, 245 (1954); No. 12, **50**, 23, 153, 173, 228 (1954); also, *Nucleonics*, **12**, No. 7, 11–42 (1954).

thermal neutrons; this is because separated lithium-7, although expensive and at present unavailable in quantity, may ultimately be important.

8.248. The general opinion at present seems to be that sodium is, on the whole, the most suitable liquid metal for use as coolant for a reactor operating at high temperatures.* If free from oxygen, liquid sodium does not attack stainless steels, nickel, many nickel alloys, beryllium, or graphite at temperatures below 600°C (1110°F). At higher temperatures mass transfer can occur in steels, but this may perhaps be controlled by additives. Because of its fairly high melting point (98°C or 208°F), there is some possibility that sodium may solidify in the reactor cooling system; this can be avoided by jacketing the sodium lines with electrical resistance heaters. In any event, after the reactor has been in operation at high power for some time, the heat which continues to be liberated after shut-down will keep the sodium in the liquid state for several days. One of the reasons why NaK, which is inferior to sodium as a heat-transfer medium, was chosen as coolant for the Experimental Breeder Reactor (EBR) was the fact that it is liquid at ordinary temperatures, so that there is no danger of solidification either inside or outside of the reactor. Also, in a fast-neutron reactor, as is the EBR, it is advantageous to replace some of the sodium by an element of higher mass number.

8.249. A drawback associated with the use of sodium as a reactor coolant, either alone or as an alloy, is the formation of sodium-24 as a result of neutron capture. This radioisotope has a half life of nearly 15 hr and emits, in addition to beta particles, two gamma-ray photons of fairly high energy (1.38 and 2.75 Mev). Consequently some shielding is necessary for coolant tanks, piping, pumps, and heat exchanger, and maintenance problems are increased. In the EBR the volume to be shielded is reduced by means of a secondary cooling circuit of sodium-potassium alloy. The exchanger in which heat is transferred from the primary to the secondary coolant is shielded, but the remainder of the secondary circuit, including the heat-exchanger (or boiler) in which steam is produced, is not radioactive and does not require shielding.

8.250. An objection which has been raised against the use of liquid sodium (or an alloy) as a coolant is the danger of fire and explosion resulting from access of air or water. Extensive studies made at Knolls Atomic Power Laboratory have established the fact that, by the exercise of proper precautions, the hazards can be greatly minimized. Confidence in the ability to handle hot liquid sodium is so great that it is planned to use it as primary coolant for the Submarine Intermediate Reactor. The heat will be transferred to water, in order to produce steam, in a heat exchanger in which mercury acts as an intermediary fluid. Thus the sodium will not come in contact with the water if the heat exchanger develops a leak, and the presence of such a leak will be readily detected from observation of the mercury.

* This statement should not be taken to imply that liquid sodium is regarded as the best coolant; it is probably the best *liquid metal* coolant now available (cf. R. F. Koenig and S. R. Vandenberg, *Metal Prog.*, **61**, No. 3, 71 (1952)).

8.251. The problem of pumping liquid metals would appear to be difficult of solution, but it has been overcome in several ways. In the so-called "electromagnetic" pumps, for example, advantage is taken of the electrical conductivity of a liquid metal to force it to flow in a pipe under the influence of a magnetic field. These pumps operate on the same principle as an electric motor. If a conductor in a magnetic field carries a current flowing at right angles to the direction of the field, a force is exerted on the conductor in a direction perpendicular to both the field and the current. In the electromagnetic pump the liquid metal is the conductor which passes through a duct located between the poles of an electromagnet (Fig. 8.1.6) A current from an external source is then passed

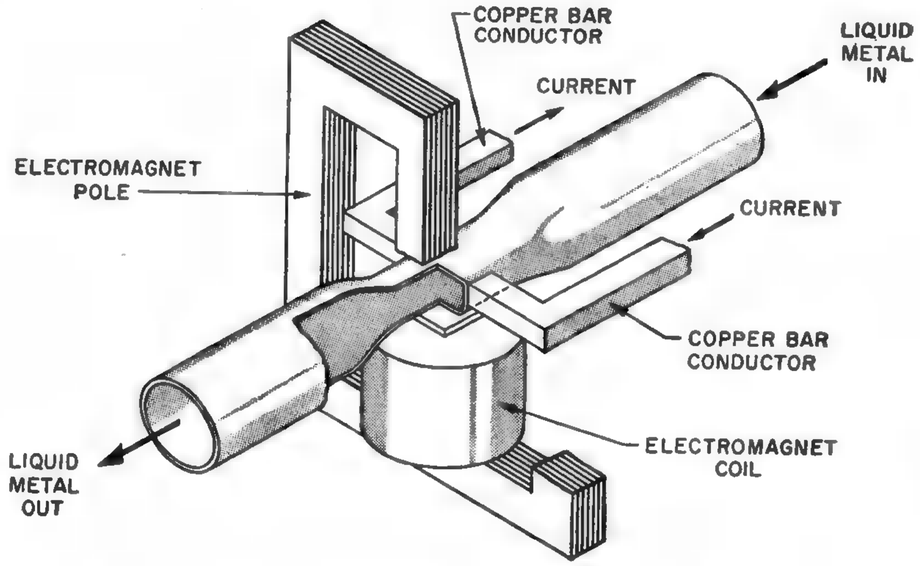


FIG. 8.16. Schematic representation of electromagnetic pump (T. Trocki)

across the liquid metal in a direction perpendicular to both the magnetic field and the desired direction of flow of the liquid metal. The force exerted on the conductor by the magnetic field then causes flow to take place. One advantage of the electromagnetic pump is that it has no moving parts, so that maintenance is greatly simplified. This is important, not only because of the nature of the liquid being pumped but also because the pump may be contaminated with radioactive material, e.g., sodium-24.*

FUSED SALTS AND HYDROXIDES AS COOLANTS

8.252. A mixture of sodium nitrite and sodium and potassium nitrates known as HTS ("heat transfer salt"), melting at 145°C (293°F), has been used for some

* For descriptions of electromagnetic and other pumps, see "Liquid-Metals Handbook," p. 245; also A. H. Barnes, *et al.*, Reports AECD-3430, AECD-3431; *Nucleonics*, **11**, No. 1, 16 (1953); J. F. Cage, Jr., Report AECU-2282; *Mech. Eng.*, **75**, 467 (1953); E. F. Brill, *ibid.*, **75**, 369 (1953).

time in industry as a heat-transfer fluid; the feasibility of molten salts as coolants on a large scale has thus been demonstrated. However, this particular mixture absorbs neutrons too readily to be of any value as a primary reactor coolant, although it might be used in a secondary system. The heat-transfer properties of fused salts are, in general, inferior to those of liquid metals. Indeed, as reactor coolants fused salts and hydroxides appear to offer no particular advantages over liquid metals, except a possible decrease of hazard and improved moderating properties. Some attention has, nevertheless, been given to fused salts and fused hydroxides as reactor components.

8.253. The fused salts proposed as coolants for high-temperature reactors are, in general, mixtures of lithium-7, sodium, and potassium fluorides, with the possible addition of fluorides of other elements having small neutron-capture cross sections. One of these is beryllium fluoride, which would contribute to the neutron moderation in a thermal reactor, if the resulting increase in viscosity could be tolerated. The fluorides appear to be stable at high temperatures and in intense radiation fields. Their heat-transfer properties are not particularly good, but they can be pumped readily. The molten fluorides can be contained, at least for a few thousand hours, at high temperatures in equipment constructed of certain nickel alloys.

8.254. The interest in fused hydroxides as reactor coolants is partly connected with the fact that the hydrogen and, to a lesser extent, the oxygen present would serve as moderators. Sodium hydroxide (melting point 318°C or 604°F) is the only hydroxide that has been considered at all seriously.* It is stable at elevated temperatures, although relatively little is known of its behavior under the influence of radiation. The problem of containing molten sodium hydroxide at temperatures above 550°C (1020°F) appears to be very difficult; below this temperature nickel and possibly certain nickel alloys show some promise.

ORGANIC LIQUIDS AS COOLANTS

8.255. In conclusion, some reference may be made to the possible use of organic liquids as reactor coolants. An organic compound consisting largely (or entirely) of carbon and hydrogen could be as good a moderator as water. If, in addition, it has a high boiling point, it would have the advantage of requiring a lower pressure than water when used as the primary coolant for a power-reactor system. Further, organic compounds, especially hydrocarbons, are on the whole very much less corrosive than water at high temperatures. A possible example of an organic moderator-coolant might be the eutectic mixture of diphenyl and diphenylene oxide, called Dowtherm A, which is used as a heat-transfer agent in industry. It melts at 25°C (78°F) and boils at 252°C (486°F) at atmospheric pressure and is thermally stable over this temperature range.

8.256. The main difficulty in connection with the use of organic compounds

* See, for example, E. M. Simons and J. H. Stang, *Chem. Eng. Prog. Symposium Series*, No. 11, **50**, 139 (1954).

as reactor coolants would appear to be their relative instability to radiation. However, some indication that this problem has been solved, at least partially, is to be found in the announcement made early in 1955 by the A.E.C. that work on the use of hydrocarbons as moderator and coolant has reached an advanced state of development. It is felt that a reactor experiment with an organic liquid as moderator-coolant may soon be justified.

8.257. Apart from the application of organic liquids as primary coolants, there may be some scope for their utilization in a secondary capacity in regions where the radiation intensity is small. For example, when sodium (or NaK) is the primary coolant, the induced radioactivity makes an intermediate heat-exchange system very desirable. An organic liquid, which did not react chemically with either sodium or water, might have definite usefulness as a secondary coolant in such a system. The relative instability to radiation would not be very serious, because the total beta and gamma radiations from the sodium-24 would be very much less than the intensity within the reactor.

Symbols Used in Chapter VIII

D	diffusion coefficient of thermal neutrons
F	feed rate of gas in diffusion cascade
L	diffusion length of thermal neutrons
L	interstage flow rate in diffusion cascade
L_n	value of L at nth stage
L_{ideal}	interstage flow rate in ideal cascade
M	migration length of thermal neutrons
M	molecular weight
N	number of atoms (or molecules) per cm^3
N_{ideal}	total number of stages in ideal cascade
N_{min}	minimum total number of stages (total reflux)
n	stage number in diffusion cascade
n_{ideal}	number of enrichment stages in ideal cascade
n_{min}	minimum number of stages (total reflux)
P	product withdrawal rate (at top of cascade)
W	waste withdrawal rate (at bottom of cascade)
x	fraction of lighter isotope in depleted (downflowing) stream
x_n	value of x at nth stage
x_f	fraction of lighter isotope in feed
x_p	fraction of lighter isotope in product
x_w	fraction of lighter isotope in waste
y	fraction of lighter isotope in enriched (upflowing) stream
y_n	value of y at nth stage
α	isotope separation factor
α^*	theoretical separation factor
α_n	separation factor at nth stage

θ	"cut" at diffusion barrier
Σ_a	macroscopic absorption cross section
Σ_s	macroscopic scattering cross section
σ_a	microscopic absorption cross section
σ_s	microscopic scattering cross section
τ	Fermi age of thermal neutrons
ξ	average logarithmic energy decrement per collision

PROBLEMS

1. In an ideal gaseous-diffusion cascade using natural uranium hexafluoride as the feed material the separation factor may be taken as 1.0043 at all stages. The fraction of uranium-235 in the product is 0.85 and that in the waste is 0.0030. Assuming the product is removed at the rate of P pounds per day, determine (*a*) the number of enrichment stages, (*b*) the number of stripping stages, (*c*) the feed rate, (*d*) the percentage recovery of uranium-235, and (*e*) the interstage flow rate at the feed stage.

2. For the conditions of the preceding problem, plot the variation of the concentration of uranium-235 and of the interstage flow rate with the stage number in an ideal gaseous-diffusion cascade (cf. Fig. 8.9). What is the total interstage flow rate for all stages of the cascade?

3. Propose a squared-off cascade that would be a good practical substitute for the ideal cascade of the two preceding problems. Estimate the total interstage flow rate and the number of stages in the enrichment and stripping sections.

4. An ion-chamber is to be placed near a reactor where the thermal flux is about 10^{11} neutrons/(cm^2)(sec). It is desired that the instrument should not need replacement for at least two years; would polystyrene be a satisfactory insulating material? Brass is frequently used for encasing ion-chambers; why would this not be suitable in the present case? Suggest alternative materials that might be used.

5. A reactor component of stainless steel (density 7.80 g/cm^3) has the following composition in weight percentages: chromium 17.5, nickel 11.0, manganese 1.0, columbium (niobium) 0.75, carbon 0.065, the remainder being iron. What is the total rate of neutron absorption per sec per cm^3 of the steel when located in a thermal flux of 10^{12} neutrons/(cm^2)(sec)? What volume of a zirconium-tin alloy (density 6.50 g/cm^3), containing 2.5 per cent by weight of tin, would capture neutrons at the same rate?

6. Determine the rate of gamma-ray energy emission (in Mev per sec per cm^3) by the steel component referred to in the preceding problem (*a*) 1 hr, (*b*) 1 day, after removal from the reactor following a long exposure. (Only those isotopic species and radiations mentioned in Table 8.13 need be considered.)

7. Cooling water, containing 5 ppm of sodium by weight, is circulated through a reactor so that in each cycle the water spends 5 sec in the reactor and 30 min in the external heat exchanger, hold-up tanks, etc. If the average thermal flux of the reactor is 10^{14} neutrons/(cm^2)(sec), calculate the equilibrium value, attained after extended operation, of the activity of the water leaving the reactor, expressed in curie/cm^3, due to sodium-24 (half life 15 hr).

*Chapter IX**

RADIATION PROTECTION†

RADIATION HAZARDS AND HEALTH PHYSICS

Radiation Problems in Reactor Design and Operation

9.1. Shortly after the discovery of X-rays and of radioactivity, toward the end of the nineteenth century, it was recognized that nuclear radiations can produce harmful effects on the living organism. Partly as a result of ignorance and partly due to accidental circumstances, a number of cases of injury, ranging from minor skin lesions to bone sarcoma and untimely deaths, were reported among radiologists and others who were exposed to excessive amounts of nuclear radiations. It was not until the early 1920s, however, that organized efforts were made to recommend safety measures to be used in the manipulation of X-rays and radium. About ten years later, maximum permissible levels of exposure to radiation were proposed, and their general acceptance led to a marked decrease in the incidence of radiation injury.

9.2. When plans were made to build the first nuclear reactor, it was realized that radiation hazards of a hitherto unimagined order of magnitude would be involved and that special precautions would have to be taken to protect the operating personnel. Consequently there was a great expansion of the field of *health physics*, the basic purpose being to devise methods for the prevention of overexposure of personnel to nuclear radiations. The efforts of health physicists have been so successful that injuries due to radiation have been virtually nonexistent, in spite of the very high levels of activity associated with reactor operation, the treatment of spent fuel, and the handling of fission products.

9.3. Radiation exposure may arise from sources external to the body or from radioactive material that has entered the body, either by inhalation, by ingestion, or by absorption through the skin. Since there is no bodily sensation to indicate the presence of nuclear radiation, instruments must be used to determine where the hazard exists. If the source of radiation is outside the body, the exposure may be reduced by shielding the source, e.g., a reactor, by limiting the time of exposure, and by using various types of remote-control devices to permit an increase in the distance between the source and the individual. Internal exposure may be controlled by rigorous cleanliness, avoidance of direct handling of active material, and proper ventilation to prevent inhalation of radioactive gases, dust, etc.

* Material contributed by E. E. Anderson, D. M. Davis, S. Glasstone, K. Z. Morgan, and F. Western.

Reviewed by E. E. Anderson, D. M. Davis, M. F. Fair, K. Z. Morgan, C. P. Straub, and H. P. Yockey.

† For reviews see Health Physics Training Lectures, 1948–1949, Report AECU-817; Health Physics Insurance Seminar, Report TID-388, 1951; K. Z. Morgan, Reports AECD-2217 and TID-5031, p. 176, 1951.

9.4. It is impossible, in spite of adequate shielding, to reduce the intensity of neutrons and gamma rays outside the shield to zero during the operation of a reactor. Further, maintenance and repair of the reactor and its equipment will involve the possibility of exposure to radiation. The coolant will usually have some induced activity after passage through the reactor, and, if this is recycled, the piping, pumps, tanks, etc., represent sources of radiation exposure for operating personnel. Even if the coolant is discharged after a single pass, e.g., air or water, its discharge must be controlled in order to avoid the harmful effects of radioactivity.

9.5. The spent fuel elements discharged from a reactor are, of course, highly active, and their treatment requires great care. Although they are usually allowed to "cool" for several days after their removal from the reactor, the fuel elements are still very radioactive when they are dissolved in the chemical processing plants. Thus the process solutions represent a serious radiation hazard. In addition, as indicated in Chapter VII, the various waste products must be disposed of in such a manner as not to jeopardize the health of the surrounding population.

9.6. Apart from the radiation hazards associated with the operation of a reactor and the chemical processing plant, it must be remembered that the fuel is radioactive, as also is the product, e.g., plutonium-239, of a converter or breeder reactor. In addition, precautions must be taken in handling material that has been placed in the reactor for irradiation. Dust in a reactor atmosphere is likely to have induced activity, as also is the air pushed out by control rods. All the foregoing considerations apply to normal reactor plant operation and maintenance. There is, however, always the possibility of a major or minor accident. Adequate plans for the protection of personnel from radiation injury must take such factors into consideration.*

9.7. It can be seen that the necessity for controlling the radiation hazard, in order to protect both operating personnel and those living in surrounding areas, places certain restrictions upon the design and operation of a reactor and its associated plant. It is in this respect, in particular, that reactor engineering differs from other branches of engineering. Some of the special aspects of

* For a description of the procedures adopted in connection with the unusual accident to the Canadian (NRX) reactor, see F. W. Gilbert, *Chem. Eng. Prog.*, **50**, 207 (1954).

radiation control which must be taken into consideration are summarized below.*

(1) The site selected for a reactor is subject to more stringent requirements than is the case for a conventional power plant, because of the possible spread of radioactivity in the event of an accident. The A.E.C. Advisory Committee on Reactor Safeguards has recommended that the radius in miles of the exclusion area for a reactor should be at least $0.01\sqrt{P}$, where P is the reactor (heat) power in kilowatts.

Actually the formula given above, based to some extent upon security considerations, is intended as a guide in determining the exclusion areas for research and development reactors, in particular. It may be modified to fit any particular situation and there are several factors which might justify a decrease in the exclusion area.† One of these is the inherent safety and reliability of the reactor, and another is a low fission-product inventory in the core. A third factor is the use of special construction that would minimize the spread of radioactivity in the event of an accident. For example, the Submarine Intermediate Reactor, at the Knolls Atomic Power Laboratory, West Milton, N. Y., is located in a large gastight, steel sphere, and the Pressurized Water Reactor, at Shippingport, Pa., is in an underground chamber.‡

(2) The size and weight of a reactor are largely determined by the amount of shielding necessary to protect operating personnel. For many reactors, such as the Experimental Breeder Reactor (EBR), for example, the shielding greatly exceeds the reactor core in both size and weight. This matter could be very significant in the design of mobile reactor power plants.

(3) Consideration must be given to the possibility of leakage of water or air with induced activity or of even small amounts of fission products into the surrounding plant. This requirement places special restrictions upon the design of equipment.

(4) The operation of the reactor may be limited by the ability to dispose of radioactive effluents, notably coolants in single-pass systems. A high-power, air-cooled reactor, e.g., located near a populated area, might have to decrease its operating power if the meteorological conditions indicated that the air could not be discharged from a high stack without representing some degree of radiological hazard on the ground. Similarly, where water is used as the coolant, it is generally stored before discharge to permit appreciable decay of induced activities, e.g., due to sodium-24. The available storage space for the water may limit the power output of the reactor.

(5) As seen in Chapter VII, the design of radiochemical plants for processing

* For review of administrative problems in radiation protection, see I. R. Tabershaw and S. J. Harris, *Nucleonics*, **12**, No. 12, 8 (1954).

† See T. Stern, *Chem. Eng. Prog. Symposium Series*, No. 11, **50**, 178 (1954).

‡ For aspects of the problem of reactor location, see L. C. Widdoes, *ibid.*, No. 11, **50**, 105 (1954); H. Hurwitz, Jr., *Nucleonics*, **12**, No. 3, 57 (1954).

spent fuel must take into consideration the necessity for the cooling period to permit decay of the shorter-lived fission products, including iodine-131. In addition there are special problems of maintenance, shielding, air filters, stacks, etc., determined by the radioactivity of the materials being handled.

(6) The radioactive nature of the waste materials, especially in processing plants, makes the problem of disposal very much more difficult than is the case in normal chemical plant operation.

Health Physics Activities

9.8. The prime function of the health physics organization is, of course, to safeguard the health of all individuals whose work is likely to involve some exposure to nuclear radiations by taking all steps that are considered necessary to minimize such exposure. In addition there is the responsibility of making sure that nothing escaping from the nuclear plant, even in the event of an accident, would represent a radiological hazard to the population of the surrounding area. The achievement of these objectives involves a number of different activities, which may be considered as falling into two broad categories: the first includes advisory and research activities as they affect the over-all nuclear energy program, and the second is concerned with the activities of the health physics group at a particular plant. These will be considered in turn.

9.9. In order to establish and maintain quantitative standards of radiation protection, health physicists together with radiologists and other scientists are members of the International Commission on Radiological Protection and the National Committee on Radiation Protection. These bodies recommend maximum radiation exposure limits for personnel, maximum permissible concentrations of certain radioisotopes in air and water, and maximum permissible amounts of such isotopes that may accumulate in the human body. Such recommendations are subject to regular review as increasing knowledge is gained of the effects of nuclear radiation on the living organism.

9.10. Continuous studies are being made of the biological effects of external radiation and of the absorption of radioactive materials by the body, so that the maximum permissible radiation dosages may be realistic. In other words, these dosages should be set at such low levels that daily exposure over many years is unlikely to cause any injury. On the other hand, the levels should not be so low as to make operation of a plant impossible. With increasing knowledge of the effects of radiation on the human body, the maximum permissible dose may be raised in an emergency or it may have to be lowered for regular exposures.

9.11. Another of the over-all advisory activities of health physicists is in connection with the choice of suitable locations for reactors and chemical processing plants. Due consideration is paid to such factors as population density, the suitability of the geological formation for underground and surface waste disposal, and the effect of local meteorological conditions on the dispersal of

airborne radioactivity. Advice is also given concerning the adequacy of shielding, remote-control handling equipment, filters in stacks, and other devices built into the reactor system for the protection of personnel. The design of so-called "hot laboratories," for experimental work with highly radioactive materials, is carried out with the advice of health physicists, so as to insure incorporation of all features necessary to prevent overexposure of individuals working in such laboratories.

9.12. Turning next to the functions of the health physics organization at a particular reactor or processing plant, one of the main responsibilities is concerned with what is called *radiation monitoring;* this involves, broadly speaking, the determination and recording of radiation dosages and dose rates at numerous locations. It will be seen later that the radiation dosage is measured in terms of the energy absorbed from the radiation, and the dose rate is the time-rate at which such energy is absorbed. In general, the total dose (or dosage) received is the product of the dose rate and the exposure time.

9.13. For the purpose of determining radiation exposures, radiation detection instruments of various kinds must be maintained in good operating condition. All persons likely to be exposed to radiation are provided with meters whereby the dose received can be measured, and any individual who has received an appreciable dose on a given day is notified so that appropriate steps may be taken to avoid overexposure. In special cases it may be necessary to measure the radioactivity present in the thyroid gland, due to accidental absorption of radioiodine, and analysis of urine and feces may be required to find out if certain radioactive species are accumulating in the body.

9.14. All accessible areas around a reactor and its associated equipment and laboratories are surveyed at frequent intervals to determine the radioactive contamination of surfaces and the amount of activity in the air. As a result of such measurements the health physicist may recommend the adoption of suitable protective actions. For example, protective clothing may be deemed necessary when the contamination is excessive. When appreciable amounts of radioactive dust are present in the air, the use of masks may be recommended. In special cases additional shielding or the employment of remote-handling devices may be required. Advice will also be given concerning the permissible working time in an area of unusually high activity. A regular survey must be made of the radioactivity of the solid, liquid, or gaseous materials discharged from the plant so as to make certain they will not constitute an environmental hazard.

BIOLOGICAL EFFECTS OF RADIATION

Acute and Chronic Exposure

9.15. The nature and extent of the injuries caused by radiation depend on various circumstances. Considering, first, sources external to the body, the exposure may be either acute or chronic. *Acute exposure,* which refers to the receipt of a relatively large dose of radiation within a short interval of time, is

highly improbable in A.E.C. projects under ordinary conditions, provided reasonable precautions are taken. It could arise through negligence, e.g., as a result of an individual walking into a beam of radiation coming through an opening in a reactor or by foolhardy actions which are contrary to standard operating procedures. A destructive accident, involving the spread of highly radioactive material, could also result in an acute overexposure to radiation. However, since such incidents have been extremely rare during more than ten years of reactor operation, it is not necessary to consider the matter of acute exposure in further detail here. Some reference to the probable effects of various acute radiation dosages will be made later (§ 9.78).

9.16. The term *chronic exposure* is used to describe the frequent, e.g., daily, exposure to relatively low radiation intensities, such as might be experienced in various operations connected with nuclear reactors. In general, for the same total radiation dosage, chronic exposure has much less serious consequences than does acute exposure. Thus a certain dose of radiation received by an individual in a few seconds may cause harm, whereas the same dose spread over a period of years may produce no noticeable effect. In other words, both the total radiation dose and the dose rate, i.e., the amount received in a given time, are important in determining the extent of biological damage, if any. The smaller the total dose and the dose rate, the less will be the injury. This is because the body evidently has certain powers of recovery so that in many cases much of the damage is not cumulative over an extended period of time. However, there are certain exceptions, notably genetic effects, where the damage is believed to be cumulative.

9.17. Most of the effects of radiation are believed to be of the so-called *threshold type;* that is to say, the dose received must exceed a certain amount before any injury occurs. This is in agreement with the fact that all human beings are continually exposed to nuclear radiation without any obvious harmful consequences. Mention may be made, in this connection, of cosmic rays, of radioactive material that is always present in air and water,* and of the radioisotopes carbon-14 and potassium-40 that are present in the body. If the dose exceeds the threshold, the damage may be reversible or irreversible; in the former case complete recovery of function is possible, whereas in the latter case the injury seems to be permanent, e.g., shortening of life span, genetic effects, or scar formation. Skin and bone-marrow damage due to low chronic exposure to radiation, for example, are reversible to some extent, but damage to brain and kidney appears to be largely irreversible.

Partial and Whole-body Exposure

9.18. The injury caused by radiation depends on the area (or volume) of the body which is exposed; in this connection a quantity called the integral dose, defined as the product of the dose and the mass of the body receiving that dose,

* The drinking water in New York City is reported to contain 5×10^{-10} microcuries (μc) of radium per cm^3; in some mineral springs the amount is as high as 10^{-7} μc/cm^3.

is often used. When X-rays or radioisotopes are used for diagnosis or treatment, limited portions of the body may receive large doses of radiation, so that the integral dose is not excessive. This may result in considerable biological damage to the irradiated area, but the over-all health of the individual is apparently not seriously affected. The exposure of the whole body or a large portion of the body to the same radiation might well be fatal. In the latter case the radiation would, of course, be absorbed by all parts of the body, and many organs might be affected in various degrees.

9.19. Different portions of the body show different sensitivities to radiation. Although there are undoubtedly variations of degree among individuals, the lymphoid tissue, bone marrow, organs of reproduction, and the gastrointestinal tract are among the most radiosensitive tissues. Of intermediate sensitivity are the skin, lungs, and liver, whereas the muscles and full-grown bones are the least sensitive.

Effects of Different Types of Radiation

9.20. Ultimately, the biological effects of nuclear radiation can be largely attributed to ionization which causes the destruction of various molecules, e.g., proteins, that play an important part in the functioning of living cells. It might therefore appear at first sight that the extent of injury would be determined by the amount of ionization produced and, hence, by the energy absorbed from the radiation. However, this is only partially true since the specific ionization, i.e., the number of ion pairs per centimeter of path (§ 2.27), has a considerable influence. In general, the greater the specific ionization, the greater the damage for a given energy absorption. For this reason, if an alpha emitter is taken into the body, it produces considerable injury. Not only is the alpha particle released inside the body, perhaps within sensitive tissue, but its energy is dissipated and absorbed within a short distance. Some indication of the relative effectiveness of different types of nuclear radiation will be given below (§ 9.57).

9.21. Although neutrons do not produce ionization directly, they are able to cause considerable biological damage as a result of secondary effects. With slow neutrons these effects are due almost entirely to the capture of the neutrons by hydrogen and nitrogen nuclei. The (n, γ) capture of neutrons by hydrogen is accompanied by the emission of 2.2-Mev photons, some of which irradiate the surrounding tissue whereas some escape from the body. The (n, p) reaction with nitrogen results in the production of protons which, like alpha particles, dissipate their energy in short paths of high specific ionization. Slow neutrons also undergo reaction with various other nuclei present in tissue, but these are believed to be of negligible significance.

9.22. When the body is exposed to fast neutrons, the particles lose their energy mainly as a result of elastic collisions with atoms of hydrogen, oxygen, carbon, and nitrogen present in living tissue, the first of these being the most important. In the collision the struck atom acquires kinetic energy which is

dissipated by ionization, excitation, and elastic collisions with other atoms. Fast neutrons can undergo the (n, p) reaction with nitrogen-14, mentioned above, and if the energy is in excess of about 1.5 Mev, the (n, α) process also occurs, so that protons and alpha particles, respectively, are produced. In addition, for energies less than 10^4 ev, the (n, γ) reaction with hydrogen becomes noticeable. The recoil atoms and the protons and alpha particles lose their energy within a very short distance of the point at which a fast neutron interacts with a nucleus. However, in considering the volume in which injury due to fast neutrons may occur, it should be remembered that such neutrons are able to penetrate considerable distances into the body.

9.23. In referring above to the biological damage caused by alpha particles, it was indicated that the alpha emitter must be in the body. Normally, the range of these nuclear radiations in tissue is so short that alpha particles of energy less than about 7.5 Mev are unable to penetrate the outer protective layer of the skin (epidermis). This means that, with only a few rare exceptions, radioactive species which emit only alpha particles do not represent an external hazard. Although beta particles have a longer range, only the most energetic are able to travel more than a few millimeters in tissue. Nevertheless, if close to (or on) the skin, beta emitters are capable of producing serious burns. It must be remembered, in any event, that both alpha and beta activity is frequently accompanied by the emission of gamma rays which are able to penetrate considerable distances into the body.

Exposure to Radiation Originating Within the Body

9.24. There is no reason to suppose that the effects of exposure to radiation from radioactive materials within the body will differ from those resulting from the same kind of radiation originating outside the body, except for the consequences of differences in distribution of the radiation dosage. These differences are the inevitable result of two factors: (1) the tendency of some chemical elements to concentrate in certain types of cells or tissue, e.g., plutonium and strontium are bone seekers, i.e., they tend to concentrate in the bone, and iodine is preferentially held in the thyroid gland; and (2) the attenuation of radiation from outside the body with depth of penetration into the body tissue.

9.25. When radioactive material enters the body the emitted radiation is in a position to produce greater injury to the body tissues than if the radiation arises from an external source. In the first place, exposure to the radiation is continuous, subject only to depletion of the quantity of radioactive material in the body by physical (decay) and biological (elimination) processes. Further, the body tissues in which the injury occurs are nearer to the source of radiation and are not shielded from it by intervening materials. This is of particular importance with alpha particles and low-energy beta particles which cannot reach sensitive tissues from outside the body, but which may dissipate their entire energy within such tissues if the sources are present in the body.

General Characteristics of Radiation Injury

9.26. The observable effects of overexposure to radiation vary widely, depending upon the circumstances involved and, to some extent, upon individual susceptibility. Among the factors which influence the results are magnitude and time-distribution of the radiation dose, the geometrical distribution within the body, type of radiation, and the age of the individual or the age range through which the overexposure occurs. From the experience gained by radiologists and others, since the discovery of X-rays and radioactivity toward the end of the nineteenth century, it appears that frequent or more or less continuous exposure of adults to radiation at average rates more than a hundred times as great as that due to cosmic rays and radioactive materials in nature (§ 9.17) may be incurred for many years without observable subsequent effects on health. However, chronic exposures to radiation at rates several thousand times that from natural sources may result in serious injury if continued over sufficiently long periods of time. Possible consequences of chronic overexposure are (1) leukopenia, (2) anemia, (3) detrimental changes in tissue structure, (4) leukemia, (5) malignant tumors, (6) cataracts, and (7) increase in the average rate of genetic mutation.* Observations on small animals also suggest the possibility of some reduction in life expectancy without the occurrence of any of these conditions.

9.27. Acute overexposure of the whole body to penetrating radiation can result in latent injury contributing to the probability of the subsequent development of one or more of the effects enumerated above. If the dose received is sufficiently large there will be in addition early nausea and vomiting, decrease in the formed-element content of the blood, epilation, loss of appetite and general malaise, diarrhea, and emaciation. The most sensitive indication of the severity of an acute overexposure of the whole body to radiation is the extent to which an early decrease occurs in the number of white blood cells. The period of time which may elapse before the initial clinical symptoms develop may vary from one or two hours for nausea to two or three weeks for epilation.

TABLE 9.1. EFFECTS OF WHOLE-BODY OVEREXPOSURES TO RADIATION

Body Reaction	*Single Exposure (acute)*	*Multiple Exposures (subacute)*	*Daily Exposures (chronic)*
Death	*****	***	*
Radiation sickness	*****	**	
Blood changes	*****	****	*
Tissue reactions	*****	***	*
Sterility (early)	*****	**	
Sterility (late)	†	*****	**
Leukemia	†	***	**
Genetic changes	†	**	**

* For a discussion of the last of these effects, see H. H. Plough, *Nucleonics*, **10**, No. 8, 16 (1952).

9.28. In Table 9.1 * an attempt is made to compare the reactions resulting from three different types of whole-body overexposure, namely, single (acute) exposure, multiple (subacute) exposures, and daily (chronic) exposures. The number of stars gives a rough indication of the degree or intensity of each particular reaction; the dagger signs indicate that these effects, which are slow in developing, would not be evident because of early death. However, if the exposed individual survived the chronic overexposure, the reactions would be similar to those for subacute overexposures. Partial shielding of the body would in general decrease the reactions observed.

RADIATION DOSE UNITS †

The Roentgen

9.29. In order to express radiation dosage in a quantitative manner, it is necessary to have a suitable unit. Such a unit should be easily reproducible and should be measurable in terms of relatively simple physical quantities. Since the purpose of knowing the radiation dose is to provide some indication of the possible injury, it would be desirable to have the unit of dosage proportional to the biological damage produced. However, the factors involved in radiation injury are so complex and so little understood, that it has not been possible to devise a unit which will satisfy both physical and biological requirements. The units actually used are thus, in a sense, a compromise.

9.30. As the effect of radiation on the living organism is mainly due to ionization, an important unit is defined with reference to this property of radiation; the unit is the *roentgen* represented by the symbol r. It is defined as "that quantity of X- or gamma radiation such that the associated corpuscular emission per 0.001293 gram of air produces, in air, ions carrying 1 electrostatic unit (1 esu) of quantity of electricity of either sign." The mass of air referred to in the definition is that of 1 cm^3 of dry air at 0°C and 760 mm Hg (1 atm) pressure, i.e., at standard temperature and pressure (STP). It should be noted that the roentgen applies only to X- and gamma radiation in air. The extension of the dose unit to other kinds of nuclear radiation will be considered later.

9.31. In order to appreciate the physical significance of the roentgen, consider 1 cm^3 of dry air at STP exposed to X- or gamma rays. As a result of the interaction of the radiation with the oxygen and nitrogen in the air, there will be produced a number of Compton recoil electrons, photoelectrons, and positron-electron pairs, in various proportions depending on the energy of the radiation (see Chapter II). These secondary particles, i.e., positive and negative electrons, will produce ion-pairs as they travel through the air. When 1 r of X- or

* "The Inservice Training Course in Radiological Health," School of Public Health, University of Michigan, 1951, p. 60 (by T. C. Evans).

† For review see R. D. Evans, *Nucleonics*, **1**, No. 2, 32 (1947); also J. H. Lawrence and J. G. Hamilton (Eds.), "Advances in Biological and Medical Physics," Vol. I, Academic Press, Inc., 1948, pp. 170–189 (by R. D. Evans); H. M. Parker, *Radiology*, **63**, 629 (1954).

gamma radiation has been absorbed by the 1 cm³ of dry air at STP, the total charge on all the ions of either sign produced would amount to 1 esu.

9.32. It is important to understand that the ion-pairs under consideration are not all formed in the 1 cm³ of air. The secondary particles originate in this quantity of air, but, since they have a considerable range, the ion-pairs that they produce, and which are presumed to be collected, will extend over a much larger volume. This fact is of importance in connection with the methods which have been devised for measurement of X- and gamma-radiation dosage in roentgens.

9.33. It may be pointed out that the radiation dose expressed in roentgens does not depend on the time during which it is received. The rate of absorption (or dosage rate) is stated in terms of roentgens per unit time, e.g., roentgens per hour or milliroentgens per hour (r/hr or mr/hr). The integrated product of the dosage rate and the exposure time gives the total dose received in roentgens.

9.34. The dose rate is frequently used as a measure of the radiation intensity or flux in a certain region. However, this is justifiable only for gamma rays of a specified energy. The radiation intensity is the rate at which the energy flows past a unit area at a given location, but the dosage rate in roentgens per unit time is a measure of the rate at which energy is *absorbed* in air at that point. Thus the dose rate will be determined by the absorption coefficient of air, and this varies with the energy of the radiation (§ 9.44).

Determination of the Roentgen

9.35. Imagine a large quantity of air exposed to a uniform radiation field, and consider a small volume at the center. The secondary particles formed by absorption of radiation in the small central volume will produce ion-pairs, many of which will be outside this volume. On the other hand, some of the secondary particles formed in other parts of the irradiated air will produce ion-pairs within the central volume. It can be shown that, provided the total volume of air is sufficiently large, the over-all number of ion-pairs formed within the small central volume is equal to the number produced by the secondary particles resulting from the interaction of the radiation with the air in the central volume. Consequently, if the ion-pairs formed in the central volume are collected, the total charge of either sign is a measure of the number of roentgens absorbed in that small volume of air. This principle, with some modification, is used in the so-called "free-air" ionization chamber, which is the primary standard for the determination of radiation dose in roentgens.

9.36. The free-air chamber must be so large that all the secondary particles formed in the small central volume are completely absorbed, i.e., produce all their ion-pairs, within the chamber. Such an instrument is too large for general use, and a modification, which closely approximates its behavior, has been developed. Suppose that the small central volume of air remains unaffected but that all the surrounding air is compressed by a factor of about 1000. The central volume would then be surrounded by a shell of dense air in which all the ionization phenomena would be reproduced on a thousandfold smaller scale. The number of ion-pairs formed within the small central volume at normal pressure would then be exactly the same as before since it would be traversed by the same secondary particles from the compressed air as from the free air.

9.37. The arrangement postulated above is not practical, but the hypothetical compressed air may be replaced by a solid having an equivalent absorption of X- or gamma radiation. The absorption is largely determined by the atomic number, and so the material should consist of elements with atomic numbers near 7 or 8, the values for nitrogen and oxygen, respectively. Secondary standards for measuring radiation dosage in roentgens are therefore made of approximately air-equivalent material, such as Bakelite, Lucite, or other plastic, consisting largely of carbon and oxygen. Ionization chambers of this type are called *air-wall chambers;* they are also sometimes referred to as *thimble chambers* because of their shape and small size. The response of an actual chamber is, to some extent, dependent on the energy of the X- or gamma radiation. An instrument of this kind should therefore be calibrated with a free-air chamber if it is to be used over a considerable range of energies.

Standard Gamma-ray Source: The Rhm

9.38. Most instruments used for radiation measurement, as described in Chapter V and also referred to below, neither are of the air-wall type nor do they measure the charge collected, i.e., the ion current, without amplification. Consequently if such instruments are to be used for the determination of radiation doses in roentgens (or dosage rates in roentgens per hour), they must first be calibrated. The obvious procedure would be to use a free-air or air-wall chamber for the purpose, but this is not convenient. In practice, radiation monitoring instruments are calibrated by means of a relatively constant source of gamma radiation, e.g., a quantity of radium in equilibrium with its short-lived decay products.* The radium is enclosed in a platinum capsule, of either 0.5 or 1 mm thickness, which absorbs alpha and beta radiations; the gamma-radiation dosage rate, at a specified distance, is measured with a standard instrument. The radium can then be used to calibrate other radiation instruments.

9.39. The "strength" of a gamma-radiation source for the purpose just described is frequently expressed in terms of roentgens per hour at 1 meter distance (abbreviated to *rhm*). Thus 1 rhm is the quantity of a specified gamma-ray source that produces a dosage rate of 1 r/hr in air at a distance of 1 meter. Experimental studies have shown that

* Since it is cheaper, cobalt-60 has been proposed for use as a gamma-ray standard in place of radium; however, because of its relatively short half life (5.3 yr), it has not proved to be very satisfactory for this purpose.

$$1 \text{ gram of radium} \equiv \begin{cases} 0.84 \text{ rhm in 0.5-mm Pt capsule} \\ 0.78 \text{ rhm in 1.0-mm Pt capsule.} \end{cases}$$

For other quantities of radium, the rhm values will be changed in proportion to the mass.

9.40. In using a standard radium source for calibration purposes, the source may be placed at any known distance d cm ($0.01d$ meter) from the instrument in air. The loss by absorption in this distance is small, and so the decrease in the radiation dose rate with distance is inversely proportional to d^2 (inverse square law), as will be seen later (§ 9.46). Hence, 1 gram of radium in a 0.5-mm capsule will produce a dose rate of $0.84/(0.01d)^2 = 8400/d^2$ r/hr at a distance d cm. Since the dose rate is proportional to the mass of radium, a source containing m milligrams will give a dose rate of $8400m/d^2$ mr/hr. In general, therefore,

$$\begin{matrix}\text{Dose rate at distance } d \text{ cm} \\ \text{from } m \text{ mg radium}\end{matrix} = \frac{Km}{d^2} \text{ mr/hr, where } K = \begin{cases} 8400 \text{ for 0.5-mm Pt} \\ 7800 \text{ for 1.0-mm Pt.} \end{cases}$$

The gamma-ray dose rate at any known distance from the radium source in air can thus be readily calculated and the result used to calibrate monitoring instruments.

The Roentgen and Energy Absorption

9.41. Since the formation of a single ion-pair in air requires a fairly definite average amount of energy, namely, 32.5 ev (§ 2.29), it follows that the roentgen is equivalent to the absorption by air of a specific amount of energy. The unit electrical charge, i.e., the electronic charge, is 4.80×10^{-10} esu, and this is the quantity of electricity carried by each member (positive or negative) of an ion-pair. Consequently $1/(4.80 \times 10^{-10})$, i.e., 2.08×10^9, ion-pairs are required to give a total charge of 1 esu of either sign. In other words, the absorption of 1 r of gamma radiation in 0.001293 gram of air results in the formation of 2.08×10^9 ion-pairs in air. The energy required to produce this number of ion-pairs is $(32.5)(2.08 \times 10^9) = 6.77 \times 10^{10}$ ev, i.e., 6.77×10^4 Mev or 0.108 erg, and this must be the energy absorbed in the 0.001293 gram of air. The energy absorbed per gram of air per roentgen is thus $0.108/0.001293 = 83.8$ ergs. The energy equivalents of the roentgen in a number of energy units are summarized in Table 9.2.

TABLE 9.2. EQUIVALENTS OF ONE ROENTGEN OF X- OR GAMMA RADIATION

1 esu of ion-pairs produced per cm³ of air
2.08×10^9 ion-pairs produced per cm³ of air
1.61×10^{12} ion-pairs produced per g of air
6.77×10^4 Mev absorbed per cm³ of air
5.24×10^7 Mev absorbed per g of air
83.8 ergs absorbed per g of air.

9.42. In view of these calculations the roentgen may be regarded as the dose of gamma radiation that would be accompanied by the absorption of 83.8 ergs of energy in 1 gram of dry air. However, it should be pointed out that, in substances of different atomic number and density, the amount of energy absorbed per gram per roentgen is different. Thus, if 1 gram of air were placed at a certain location in a gamma-ray field and the energy absorbed were 83.8 ergs, then 1 gram of soft tissue at the same location would absorb about 93 ergs from the radiation, provided the photon energy is not too high. In 1 gram of bone the energy absorption would be still larger, e.g., 150 ergs or more. In each case, however, the radiation dose would be 1 r. Thus the dose in roentgens, based on the definition in § 9.30, i.e., in terms of absorption in air, is independent of the medium exposed to the radiation and the amount of energy absorbed by that particular medium.

9.43. The rate of energy absorption in air of gamma radiation is determined by the *energy-absorption coefficient* for that particular energy. This quantity is not the same as the total absorption coefficient defined in § 2.60, since the latter includes the whole contribution of the Compton effect. Actually, only part of the Compton interaction leads to a dissipation of the gamma-ray energy. Consequently the energy-absorption coefficient is obtained by subtracting the purely scattering contribution from the total absorption coefficient. Suppose that at a certain location the gamma-ray flux of energy E Mev is ϕ photons/(cm²)(sec); the radiation intensity, as defined in § 9.34, is then ϕE Mev/(cm²)(sec). If μ_e cm^{-1} is the energy-absorption coefficient of air for the specified gamma radiation,* of energy E Mev, then, since the absorption coefficient is equivalent to a macroscopic cross section (§ 2.116), it follows that the rate of energy absorption in air is $\phi E\mu_e$ Mev/(cm³)(sec).

9.44. According to the data in Table 9.2, the absorption of 6.77×10^4 Mev of energy per cm³ of air is equivalent to 1 r. Hence, in the case under consideration, the dose rate is $\phi E\mu_e/(6.77 \times 10^4)$ r/sec; thus,

$$\phi \text{ photons/(cm}^2\text{)(sec)} \equiv \frac{\phi E\mu_e}{6.77 \times 10^4} \text{ r/sec.} \qquad (9.44.1)$$

This result points up the fact indicated earlier that the dose rate due to gamma radiation may vary even though the intensity is constant, if the energy is not the same. In equation (9.44.1), the intensity is equal to ϕE but the dose rate is proportional to $\phi E\mu_e$, and if the energy-absorption coefficient of air varies with energy, as it does in fact (Fig. 9.1),† the dose rate may change even though the intensity, at different energy values, is constant. By rearranging equation (9.44.1), it is readily seen that

* In the literature this coefficient is often represented by $\mu - \sigma_s$, where μ is the total (linear) absorption coefficient and σ_s is the scattering contribution (actually the *macroscopic* cross section for scattering).

† W. S. Snyder and J. L. Powell, Report AECD-2730.

$$1\ \text{r/sec} \equiv \frac{6.77 \times 10^4}{E\mu_e}\ \text{photons/(cm}^2\text{)(sec)}$$

or, in terms of more convenient units,

$$1\ \text{mr/hr} \equiv \frac{0.0188}{E\mu_e}\ \text{photons/(cm}^2\text{)(sec)}. \qquad (9.44.2)$$

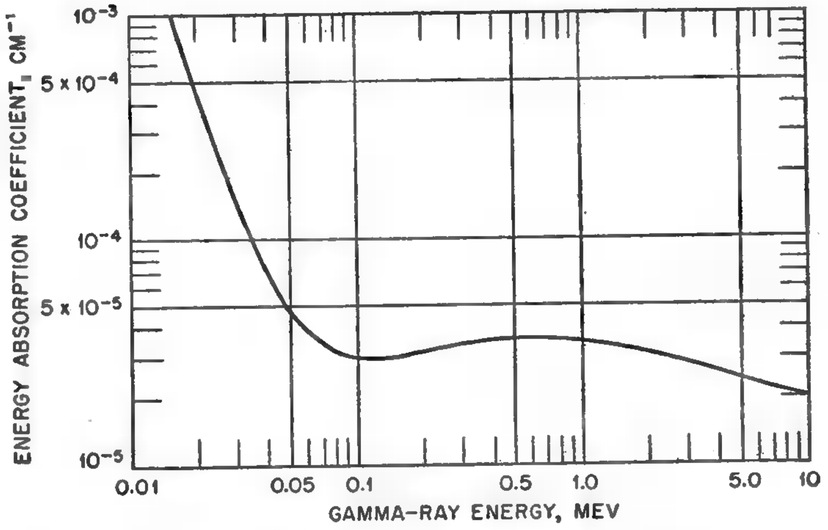


FIG. 9.1. Variation of energy-absorption coefficient of air with gamma-ray energy (W. S. Snyder and J. L. Powell)

DOSE RATE AND RADIOACTIVE SOURCE STRENGTH

9.45. An aspect of the relationship between the strength of a source of gamma radiation and the dose rate at a specified distance was given in § 9.40, and a more general treatment is of interest. Consider a radioactive *point source* having a strength of C curies (see § 2.14, *et seq.*), so that it decays at the rate of $3.7 \times 10^{10}C$ dis/sec. It will be postulated that one gamma-ray photon of energy E Mev is produced in each act of nuclear disintegration, so that the source yields $3.7 \times 10^{10}C$ photons/sec. If the radiation from the point source is emitted uniformly in all directions, then at a distance d cm from the source, the photons will be distributed uniformly over the surface of a sphere of area $4\pi d^2$ cm^2 (Fig. 9.2). Neglecting the absorption of radiation in the short distance (less than a few meters) from the source to the point of observation (§ 9.40), the gamma-ray flux at the distance d from the unshielded source is $3.7 \times 10^{10}C/4\pi d^2$ photons/(cm^2)(sec). Upon combining this result with equation (9.44.2), it is found that

$$\text{Dose rate at distance } d \text{ cm from } C \text{ curie source} = \frac{3.7 \times 10^{10}CE\mu_e}{4\pi d^2 \times 0.0188}$$

$$= 1.56 \times 10^{11}\frac{CE\mu_e}{d^2}\ \text{mr/hr}, \qquad (9.45.1)$$

the energy E being in Mev.

9.46. It will be observed that according to this equation the dose rate at a given distance is inversely proportional to the square of the distance, and so it is an expression of the inverse square law referred to earlier. Its basis, as may be seen from Fig. 9.2, is the fact that with increasing distance from the source the same number of photons (apart from a negligible amount of absorption) are spread over a surface which increases in area with the square of the distance. It is this circumstance which makes appreciable separation from a radioactive source an important factor in radiation protection.

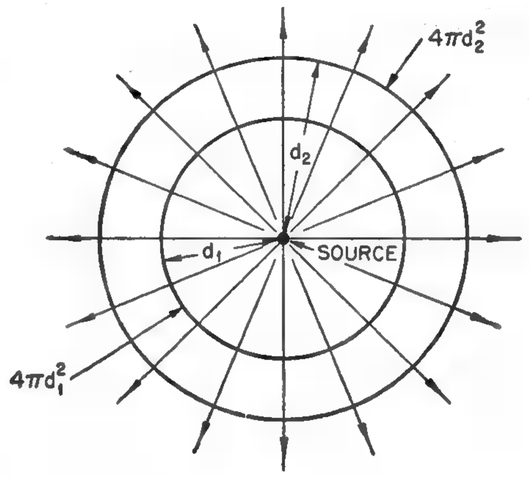


FIG. 9.2. Inverse-square law of radiation attenuation

9.47. An examination of Fig. 9.1 shows that over a considerable range of photon energies, e.g., from 0.07 to 2 Mev, the energy-absorption coefficient for air varies only between 3.0×10^{-5} to 3.7×10^{-5} cm^{-1}. For an approximate treatment, μ_e may therefore be taken to have a constant value of 3.35×10^{-5} cm^{-1}; in these circumstances equation (9.45.1) becomes

$$\text{Dose rate at distance } d \text{ cm from } C \text{ curie source} = 5.2 \times 10^6\frac{CE}{d^2}\ \text{mr/hr}. \qquad (9.47.1.)$$

It should be pointed out that the use of a constant value for μ_e makes the dose rate proportional to the radiation intensity. Since μ_e actually varies with the photon energy, this result is not exact; however, it is accurate, in the present circumstances, to within ±10 per cent.

9.48. A simple and useful form of equation (9.47.1) is obtained if the distance from the source to the observation point is expressed in feet; if this distance is f ft, then, since 1 ft = 30.5 cm, it is found that

$$\text{Dose rate at distance } f \text{ ft from } C \text{ curie source} \approx 6 \times 10^3\frac{CE}{f^2}\ \text{mr/hr}$$

$$\approx \frac{6CE}{f^2}\ \text{r/hr}.$$

It follows from this result that at a distance of 1 ft from a point gamma-ray source of strength C curies the dosage rate is approximately $6CE$ r/hr.

9.49. The foregoing expressions apply to an unshielded source, but if an absorbing material of thickness x cm and (total) absorption coefficient of μ cm^{-1} is placed between the source and the point at which the dose rate is being calculated, the general equation (9.47.1) becomes

$$\text{Dose rate at distance } d \text{ cm from } C \text{ curie source with } x \text{ cm absorber} = 5.2 \times 10^6 \frac{CE}{d^2} e^{-\mu x} \text{ mr/hr}, \quad (9.49.1)$$

where the exponential factor on the right allows for absorption of the gamma radiation (§ 2.60).

9.50. If the postulate made above that every disintegration is accompanied by one photon is not correct, the equations must be modified accordingly. In the general case, which includes mixtures of radioactive species, CE is replaced by the sum of a number of $CE\gamma$ terms, where γ is the fraction of disintegrations (accompanied by gamma rays of energy E) of the species of strength C curies. For a radium source, in equilibrium with its decay products of short life, in a platinum capsule of wall-thickness x cm, the use of the appropriate values of γ and E would make equation (9.49.1) take the form*

$$\text{Dose rate at distance } d \text{ cm from } C \text{ curies of radium in } x \text{ cm Pt capsule} \approx 9.3 \times 10^3 \frac{Ce^{-1.9x}}{d^2} \text{ r/hr},$$

where 1.9 cm^{-1} is the absorption coefficient of platinum for the gamma rays from radium. Upon taking into account the fact that 1 gram of radium has a strength of 1 curie, it can be readily shown that this result reduces to the expression in § 9.40 for the dose rate from m mg of radium.

9.51. Strictly speaking, the factor $e^{-\mu x}$ should include an allowance for absorption of gamma radiation in the d cm of air separating the source from the observation point; however, μ for air is of the order of 10^{-4} cm^{-1} (see Fig. 2.10), and so, for distances up to a few meters, $e^{-\mu x}$ is so close to unity that absorption by the air may be neglected.

Example A block of metallic cobalt, weighing 1 lb (450 g), contains 0.015 per cent of radioactive cobalt-60, as the result of neutron capture in a reactor. Calculate the dose rate that would be received at a distance of 1 yd (91.5 cm) in air. What thickness of lead would have to be interposed to reduce this to 7.5 mr/hr?

According to equation (2.15.3),

$$C = \frac{1.13 \times 10^{13} g}{A t_{\frac{1}{2}}},$$

where g is the mass of cobalt-60 in grams, i.e., $(450)(0.015 \times 10^{-2}) = 0.0675$ g; A is the atomic weight of the isotope, i.e., 60; and $t_{\frac{1}{2}}$ is its half life (5.3 yr) in seconds, i.e., 1.7×10^8 sec. Upon substituting these values, it is found that the strength of the cobalt source is 75 curies.

In its radioactive decay, cobalt-60 emits two gamma-ray photons, with energies 1.16 and 1.32 Mev, respectively, in each disintegration. Hence, in the present case, E is 2.48 Mev. With these values for C and E, and setting d equal to 91.5 cm, equation (9.47.1) gives the required dose rate as 1.15×10^5 mr/hr or 115 r/hr.

To determine the thickness of lead required to reduce the dose rate to 7.5 mr/hr, use is made of equation (2.60.2), with $I_0 = 1.15 \times 10^5$ and $I_x = 7.5$. It can be seen from Table 2.4 that μ for lead for gamma rays with energies 1.16 and 1.32 Mev is about 0.75 cm^{-1}. Hence,

* K. Z. Morgan, unpublished.

$$7.5 = 1.15 \times 10^5 e^{-0.75x},$$

so that x, the thickness of lead, is found to be about 13 cm (5.1 in.).

9.52. In deriving the equations relating the dose rate to the source strength, the source has been regarded as a point; in other words, it has been assumed that its size is small in comparison with the distance separating it from the position where the dose rate is to be estimated. In many instances, however, the source may be a relatively large sphere, a parallelepiped, a disk, or other shape that cannot be treated as a point. The equations which permit dose rates from a point source to be converted into the values to be expected from sources of other forms will be given in Chapter X.

The Rem, Rad, and Rep

9.53. As stated above, the roentgen applies strictly to X- and gamma-ray dosages, but, in view of the similar biological effects of other radiations, it is necessary to have a dosage unit which can be used for all types of nuclear radiation. One such unit is the *rep* (roentgen equivalent physical), originally defined as the dose of any nuclear (or ionizing) radiation that results in the absorption of 83 ergs/gram of tissue. In order to make the rep more nearly equivalent to the roentgen, for measuring the dose of X- or gamma radiation absorbed in soft tissue, the rep has been used in recent years to denote a dose of 93 ergs absorbed per gram of tissue. It should be noted, however, that the equivalence is not exact throughout the soft tissues of the body, particularly for X- or gamma radiation of very high energy, and fails completely for absorption in bone.

9.54. In 1953 the International Commission on Radiological Units adopted a new unit of absorbed radiation dose, called the *rad*,* which is expected to supersede the rep in due course. The rad represents 100 ergs/gram, imparted by ionizing particles to the irradiated material *at the point of interest.* Essentially the rad differs from the rep in the respect that it does not prescribe the material in which the energy is absorbed, in addition to being somewhat different in magnitude.† Further, the name rad does not imply any relationship to the roentgen, as does the rep.

9.55. Although all ionizing radiations are capable of producing the same kinds of biological effects, the absorbed dose (measured in reps or rads) which will produce a particular effect may vary considerably from one radiation to another. In general, those particles which cause higher specific ionizations along their paths are those having the greater biological effectiveness. For various reasons it is convenient to compare the biological effectiveness of ionizing radiations with that of X-rays of 200-kev energy. On this basis, a quantity called the *relative biological effectiveness* (or RBE) is defined by

* See *Nucleonics*, **12**, No. 1, 11 (1954).

† For *soft tissue* the rep and the rad differ by a few per cent only, so that they may then be taken to be essentially equal.

$$\text{RBE} = \frac{\text{Physical dose of 200-kv X-rays to produce effect of interest}}{\text{Physical dose of comparison radiation to produce same effect}}.$$

The value of the RBE for a particular radiation may depend upon several factors, e.g., the kind and degree of the biological effect, the nature of the organism or tissue, and the rate at which the dose is delivered.

9.56. It is evident that the rep and the rad, being simply indications of the amount of energy absorbed from the radiation, irrespective of the nature of the latter, do not provide an adequate measure of the resulting biological injury, since they do not take into account the relative effectiveness of the radiations. In an attempt to devise a unit which would provide a better criterion of biological injury when applied to different radiations, the *rem* (roentgen equivalent man) has been introduced. The rem is now defined by the relationship

$$\text{Dose in rems} = \text{RBE} \times \text{Dose in rads}.$$

9.57. Since, as stated above, the actual value of the RBE depends upon various circumstances, it is apparent that the equivalence implied by the term rem will apply only toward the production of the specific effect and under the conditions for which the particular RBE was determined. However, for purposes of radiation protection, it has become the practice to use a single value of the RBE for different effects of a particular kind of radiation, based on that effect for which the RBE is believed to be highest. The data in Table 9.3, except for thermal neutrons, are taken from the provisional recommendations made in 1954 by the International Commission on Radiological Protection (cf. § 9.9) for radiations of interest in nuclear reactor operation. The effects considered to be critical in the determination of the various RBE values are given in each case.

TABLE 9.3. RECOMMENDED VALUES OF RELATIVE BIOLOGICAL EFFECTIVENESS

Radiation	*RBE*	*Biological Effect*
X-rays, gamma rays, beta particles, and electrons of all energies	1	Whole body irradiation (blood-forming organs critical)
Fast neutrons and protons up to 10 Mev	10	Whole body irradiation (cataract formation critical)
Thermal neutrons	3–4*	.....
Naturally occurring alpha particles	10	Carcinogenesis outside skeleton†

* The RBE value quoted, estimated from the calculations of W. S. Snyder (*Nucleonics*, **6**, No. 2, 46 (1950)), is uncertain, because the biological effects produced by thermal neutrons depend on the variable amounts of energy expended in tissue by the gamma rays and protons arising from their capture (§ 9.21). The critical effect is probably cataract formation.

† For carcinogenesis of the skeleton, alpha-emitting isotopes are compared with radium-226, on the basis that the maximum permissible amount of radium in the skeleton is 0.1 microcurie.

9.58. Since 1 rem of one kind of radiation will (conventionally, at least) produce the same biological damage to a given tissue as 1 rem of any other kind, doses of different radiations when measured in rems are additive. This is not true, of course, from the biological standpoint, for doses expressed in reps or rads. However, by making use of the accepted RBE values the measured doses of different radiations expressed in roentgens or rads can be converted into rems and then added.

Example An individual receives, in the course of a week, a dose of 0.1 r (or 0.1 rad) of gamma radiation and 0.02 rad of fast neutrons. Determine the total dose received in rems.

For gamma rays the RBE is 1, and so 0.1 rad is equivalent to 0.1 rem. For fast neutrons the RBE is 10, so that the dose of 0.02 rad is equivalent to (10)(0.02) = 0.2 rem. The total dose received is thus 0.1 + 0.2 = 0.3 rem.

STANDARDS OF BIOLOGICAL PROTECTION

Maximum Permissible Exposure: External Sources*

9.59. For the protection of persons whose work renders them liable to radiation exposure, certain dosage maxima have been recommended. On the basis of information obtained from radiologists, X-ray technicians, etc., from the biological damage caused by radium, from animal experiments, and from the naturally occurring radiation in the body and in air and water, the International Commission on Radiological Protection recommended that the *maximum permissible exposure* (or MPE) can be set at a dose of 0.3 rem/week, i.e., 300 mrem/week, over the whole body.† This means that, as far as present knowledge goes, a person receiving 300 mrem/week over the whole body for a period of years will not suffer any detectable injury.‡ If the radiation exposure is restricted to certain parts of the body, such as the skin, for which the recovery is rapid, the MPE may be safely increased above 300 mrem/week. It should

TABLE 9.4. TOTAL MAXIMUM PERMISSIBLE EXPOSURE TO EXTERNAL SOURCES OF RADIATION

Radiation	mr/week	mrem/week	mrep/week	mrad/week (in tissue)
X or gamma	300	300	300	300
Beta particles	—	300	300	300
Thermal neutrons	—	300	~100	~100
Fast neutrons	—	300	30	30

* For a complete review, see National Bureau of Standards Handbook 59, U. S. Government Printing Office, 1954.

† *Nucleonics*, **8**, No. 1, 32 (1951).

‡ The great majority (about 95 per cent) of persons employed in A.E.C. projects receive less than 10 per cent of the MPE, according to health physics records of recent years.

be noted that the MPE given here applies to radiation from external sources; the problem of radioactive material within the body will be considered later.

9.60. The MPE is stated in terms of rems in order to allow for exposure to various types of ionizing radiations. Bearing in mind the relationship between the rem and the rep, the definition of the rad, and the RBE values for different radiations, Table 9.4 has been compiled. In view of the essential equivalence of the rad in tissue with the rep, the numbers in the last two columns are the same. If the exposure to ionizing radiations is restricted to the hands and forearms, e.g., in operations involving the handling of a beta emitter, the MPE may be increased by a factor of 5, i.e., to 1.5 rem/week.

Maximum Permissible Radiation Fluxes

Gamma-ray Flux

9.61. The flux of gamma-ray photons which would correspond to the maximum permissible exposure can be obtained very simply from equations already derived. If, as in § 9.47, the energy-absorption coefficient (μ_e) for air is taken to have the constant value of 3.35×10^{-5} cm^{-1} in the energy range from 0.07 to 2 Mev, which is correct within about 10 per cent, it is found from equation (9.44.2) that

$$1 \text{ mr/hr} \equiv \frac{5.6 \times 10^2}{E} \text{ photons/(cm}^2\text{)(sec)}.$$

The MPE for gamma radiation is 300 mr/week and so, assuming a working week of five 8-hr days, the MPE is equivalent to a dose rate of 300 mr/40 hr, i.e., 7.5 mr/hr. It follows, therefore, that, to a fair approximation,

$$\text{MPE} \equiv \frac{4000}{E} \text{ photons/(cm}^2\text{)(sec)}.$$

Thus a gamma-radiation flux of about 4000 photons/(cm^2)(sec) of energy of 1 Mev, or of 2000 photons/(cm^2)(sec) of energy of 2 Mev, would provide the maximum permissible continuous exposure for each 8-hr day of a 5-day working week. For longer exposure periods the radiation flux corresponding to the MPE must be decreased proportionately.

Fast-neutron Flux

9.62. The evaluation of the maximum permissible flux of fast neutrons is based on the view that the biological damage is due essentially to the ionization caused by recoil protons (hydrogen nuclei) in animal tissues (§ 9.22). If N is the number of hydrogen atoms/cm^3 in tissue and σ is the scattering cross section for fast neutrons of a specified energy, the rate of formation of recoils for first collisions of the incident neutrons is $N\sigma\phi$ protons/(cm^3)(sec), where ϕ neutrons/(cm^2)(sec) is the incident fast flux. On the average, a neutron loses half its

energy in a collision with a hydrogen nucleus.* Therefore the recoil protons from the first collisions dissipate energy at the rate of $\frac{1}{2}N\sigma\phi E$ Mev/(cm^3)(sec), where E is the neutron energy in Mev.

9.63. The absorption of 100 ergs, i.e., 6.2×10^7 Mev, per gram of tissue is equivalent to an exposure of 1 rad; hence, it can be readily seen that

$$\phi \text{ fast neutrons/(cm}^2\text{)(sec)} \equiv \frac{N\sigma\phi E}{(2)(6.2 \times 10^7)} \text{ rad/sec}$$

or

$$1 \text{ mrad/hr} \equiv \frac{29}{N\sigma E} \text{ fast neutrons/(cm}^2\text{)(sec)},$$

where N is now the number of hydrogen nuclei (or atoms) *per gram* of tissue, i.e., about 6.0×10^{22}. The RBE of fast neutrons is taken as 10 (Table 9.3) so that 1 rad is equivalent to 10 rem; consequently,

$$1 \text{ mrem/hr} \equiv \frac{2.9}{N\sigma E} \text{ fast neutrons/(cm}^2\text{)(sec)}.$$

Using the MPE of 300 mrem/40 hr, i.e., 7.5 mrem/hr, and inserting the value given above for N, the result is

$$\text{MPE} \equiv \frac{3.6 \times 10^{-22}}{\sigma E} \text{ fast neutrons/(cm}^2\text{)(sec)}.$$

For neutrons of 2 Mev energy, σ is 3×10^{-24} cm^2 per nucleus, and so the maximum fast-neutron flux, on the basis of this calculation, is roughly 66 neutrons/(cm^2)(sec) for exposures averaging 40 hr/week.

9.64. As a result of more detailed calculations, taking into account the successive collisions the neutron may experience in the body, the effects of recoil nuclei other than protons, and other reactions, the (rounded) values recorded in Table 9.5 have been obtained for the maximum permissible flux of fast neutrons of several energies, assuming an exposure of 40 hr/week.†

TABLE 9.5. MAXIMUM PERMISSIBLE AVERAGE FAST-NEUTRON FLUX BASED ON 40 HR/WEEK EXPOSURE

Neutron Energy (Mev)	*Flux* n/(cm^2)(sec)	*Neutron Energy* (Mev)	*Flux* n/(cm^2)(sec)
10	30	1	60
5	30	0.5	80
3	30	0.1	200
2	40	0.01	1000

* The fraction used here is the arithmetic mean energy loss per collision. Since it is larger than the geometric mean of $1/e$ (§ 3.61), the final result will be a somewhat lower maximum permissible flux.

† Cf. W. S. Snyder and J. Neufeld, Report ORNL-1872; to be published in *Brit. Jour. Radiology*, 1955.

Thermal-neutron Flux

9.65. The biological effect of slow (thermal) neutrons in body tissue may be attributed essentially to the 2.2-Mev gamma rays accompanying the (n, γ) capture of neutrons by hydrogen and to the 0.57-Mev protons liberated in the (n, p) reaction with nitrogen. Although the reaction with hydrogen accounts for most of the energy released, it is the reaction with nitrogen that is responsible for the greater biological effect at the depth of tissue at which the absorbed dose is a maximum. This is the result of two circumstances: (1) the energy of a proton is absorbed very close to the point at which it is produced, whereas much of the gamma radiation is either absorbed at a considerable distance from its point of origin or it escapes from the body without absorption; and (2) the RBE of the protons is considered to be roughly ten times that of the gamma radiation (see Table 9.3).

9.66. The relation of the incident flux of thermal neutrons to the distribution of thermal-neutron interactions within the body and that of the gamma radiation resulting from the interaction with hydrogen is too complex to permit of a valid approximation to the maximum permissible thermal-neutron flux being made by elementary methods of calculation.* The provisional value recommended in 1954 by the International Commission on Radiological Protection is an average of 2000 thermal neutrons/(cm²)(sec) based on an exposure of 40 hr/week.

Summary

9.67. From the standpoint of the design of shields for nuclear reactors, the important radiations are gamma rays, fast neutrons, and thermal neutrons, which have been discussed above. The recommended average values for the maximum permissible fluxes of these radiations, assuming an external exposure of 40 hr/week, are summarized in Table 9.6. The value for beta particles, although of lesser significance, is included for the sake of completeness. Alpha particles are omitted since they are not a serious hazard outside the body.

TABLE 9.6. MAXIMUM PERMISSIBLE AVERAGE FLUXES OF EXTERNAL NUCLEAR RADIATIONS IN 40-HR/WEEK EXPOSURE

Radiation	*Energy*	*Flux*
Gamma rays	1 Mev	4000 photons/(cm²)(sec)
Beta particles	1 Mev	94 particles/(cm²)(sec)
Thermal neutrons	0.025 ev	2000 neutrons/(cm²)(sec)
Fast neutrons	2 Mev*	40 neutrons/(cm²)(sec)

* For other energies see Table 9.5.

* Cf. W. S. Snyder, *Nucleonics*, **6**, No. 2, 46 (1950).

Maximum Permissible Concentrations: Internal Sources

9.68. Since the radiation dose received from a particular radioactive material in the body is proportional to the quantity of that material present, excessive exposure to radiation from internal sources is avoided by limiting the body content of the material. This is achieved by restricting the rate at which the material enters the body. In many instances intake into the body results primarily from the occurrence of the radioactive material in the air (or water) supply. It is then desirable to know the maximum average concentration of the active species which may be permitted in the air (or water) without eventually resulting in excessive accumulation of the material in the body.

9.69. Maximum safe average body contents of most radioisotopes are not known, since there is insufficient direct experience for establishing levels at which individual isotopes, except possibly radium and radioiodine, begin to cause injury to human beings. For most of the radioactive materials which, like radium, accumulate in the skeleton, maximum safe levels for the human body are estimated, directly or indirectly, from comparative experimental studies with other species of mammals. For radioisotopes which concentrate in body organs or tissues other than the skeleton, it is assumed that the body content should not exceed the lowest level which will result in an average dose rate at some part of the body of 0.3 rem/week (cf. § 9.59).*

9.70. It may be observed that, on the basis of this criterion, if most of a particular radioisotope is fairly uniformly distributed in a single organ or tissue, the maximum permissible body content is inversely proportional to the mass of that organ or tissue. For example, since most of the iodine in the body is concentrated in the thyroid gland, the average total activity of iodine-131 permitted is much less than that of sodium-24 which is distributed throughout the body. This is the case in spite of the fact that the average energy of the radiation from iodine-131 is less than that from sodium-24.

9.71. The quantity of an active material remaining in the body after its entry is dependent upon its radioactive half life and also upon its *biological half life;* the latter is the time taken for the amount of the element in a particular body tissue to decrease to half of its initial value due to elimination by natural processes. Combination of the radioactive and biological half lives gives rise to the *effective half life*, which is the time required for the amount of a specified radioisotope to decrease to half the original value as a result of both radioactive decay and natural elimination. It is understood, of course, that there is no replenishment of the isotope from any source during the time under consideration. In

* From the standpoint of radiation protection this criterion is regarded as being conservative. There is no reason to expect greater injury to the body from irradiation of a limited portion than from irradiation of the whole body at the same rate of 0.3 rem per week, and in many cases the portion of the body involved could safely be irradiated at higher average rates. However, it is generally not possible or practical to make such distinctions.

the simplest case, the effective half life in the body as a whole is essentially that in the principal tissue in which the material concentrates.

9.72. The maximum safe average rate of entry into the body of a given radioisotope is determined by (1) initial retention and distribution in the body, (2) maximum safe average body content, and (3) effective half life. These three factors are not simple ones, since each in turn is dependent upon two or more other factors. Initial retention, for example, is not only related to the biochemical properties of the chemical element (or, in some cases, the chemical compound), but also to the mode of intake into the body. Thus, nearly all of the iodine entering the digestive tract may be initially retained in the body, whereas only about 0.01 per cent of ingested plutonium is retained. On the other hand, it is estimated that as much as 20 per cent of soluble plutonium entering the respiratory system may reach the circulatory system.

9.73. If other factors are more or less the same, the rate of intake into the body which will eventually result in a maximum permissible body content is inversely proportional to the effective half life of the radioisotope in the critical organ or tissue. If either the radioactive half life of the given isotope or the biological half life of the element in the tissue is very short compared to the other, the effective half life is approximately equal to the shorter of the two, since this essentially determines the rate of loss of the particular material from the tissue.

9.74. Of the radioactive species involved in reactor operations or in the processing of reactor fuels, plutonium-239, natural uranium (and its isotopes), natural thorium, strontium-90, and iodine-131 have low maximum permissible average concentrations, as measured in activity per unit volume, in air breathed by human beings. This is because of the importance of one or more of the three factors mentioned in § 9.72. In the case of plutonium-239, all three factors contribute to limiting the maximum permissible concentration in air to an extremely

TABLE 9.7. MAXIMUM PERMISSIBLE CONCENTRATIONS IN WATER AND AIR

Substance	Radiations	Critical Organ	MPC, $\mu c/cm^3$	
			In Water	In Air
Natural uranium (sol.)	α	Kidneys	7×10^{-5}	1.7×10^{-11}
Natural uranium (insol.)	α	Lungs	—	1.7×10^{-11}
Uranium-233 (sol.)	α	Bone	1.5×10^{-4}	1×10^{-10}
Uranium-233 (insol.)	α	Lungs	—	1.6×10^{-11}
Plutonium-239 (sol.)	α	Bone	1.5×10^{-6}	2×10^{-12}
Plutonium-239 (insol.)	α	Lungs	—	2×10^{-12}
Iodine-131*	β and γ	Thyroid	6×10^{-5}	6×10^{-9}
Strontium-90 + Yttrium-90	β	Bone	8×10^{-7}	2×10^{-10}
Strontium-89	β	Bone	7×10^{-6}	2×10^{-8}

* Recommendations of International Commission on Radiological Protection, 1954.

small value. However, because of its low uptake from the digestive tract, the maximum permissible concentration of plutonium-239 in water is about twice as great as that of strontium-90.

9.75. Some of the values of the maximum permissible concentrations (MPC) in air and water, for a few of the radioisotopes of special interest in reactor operations, are given in Table 9.7, the MPCs being expressed in microcuries per cubic centimeter ($\mu c/cm^3$).*

Acute Radiation Dosages: Accidental and Emergency Exposures

9.76. The maximum permissible average rates of exposure to radiation discussed in preceding sections apply to chronic radiation exposures continued over periods of many years. In the event of an accident or of an emergency involving unplanned conditions, persons may incur, knowingly or otherwise, large acute doses of radiation. In this connection, consideration has been given to the possibility of setting a maximum permissible dose for acute exposure under emergency conditions. However, it is generally agreed that no such limit, having real significance, can be established.

9.77. The fundamental basis of the concept of an emergency dose is that in special circumstances an individual might accept the risk of possible or certain injury from an acute radiation exposure. However, not only is the risk that would be acceptable dependent upon the degree of the emergency, but there are no grounds for correlating the magnitude of the risk and the character of the emergency. Further, in those phases of emergencies in which immediate action is required to avert serious or catastrophic results, it would rarely be possible

TABLE 9.8. PROBABLE EARLY EFFECTS OF ACUTE RADIATION EXPOSURES OVER WHOLE BODY

Acute Dose	*Probable Effect*
0 to 25 r	No detectable clinical effects.
25 to 50 r	Possible blood changes but no other clinically detectable effects.
50 to 100 r	Nausea and fatigue, blood-cell changes, and some injury, but no disability.
100 to 200 r	Nausea, vomiting, fatigue, and reduced vitality; depression of nearly all blood elements. (Recovery in nearly all cases within 3 to 6 months.)
200 to 400 r	Same as above, with immediate disability; some deaths within 2 to 6 weeks.
400 r	Fatal to 50 per cent.
600 r or more	Fatal to nearly all within 2 weeks.

* MPC values for many other radioisotopes that may be encountered in various aspects of the atomic energy field, recommended by the National Committee on Radiation Protection, are given in National Bureau of Standards Handbook 52, U. S. Government Printing Office, 1953. This handbook also contains a discussion of the considerations upon which the recommended values are based. See also, K. Z. Morgan, Report TID-5031, p. 176; K. Z. Morgan and M. R. Ford, *Nucleonics*, **12**, No. 6, 32 (1954).

to predict (or measure) in advance the radiation dose that might be received as a result of such action.

9.78. There are, however, situations in which it is desirable to estimate the probable results of acute doses of radiation, ranging from a few roentgens to several hundred roentgens (or their equivalent), received over a short period of time. Such estimates, which are summarized in Table 9.8, are based upon limited data and must be regarded as giving only an approximate indication of the early effects of whole body exposure for average persons. Other aspects of radiation injury resulting from acute overexposures were mentioned in § 9.26.

PROTECTION OF PERSONNEL*

Remote Control

9.79. The protection of personnel from radiation requires the design of suitable shielding for the reactor and for all vessels, piping, pumps, etc., which contain or carry radioactive material. In addition, measures must be taken to prevent exposure to the radiations from spent fuel elements after removal from the reactor. Some aspects of the shielding problem will be considered in the next chapter, but, in general, it may be stated that adequate shielding requires the use of considerable thicknesses of material, e.g., several feet of concrete or of water. This means that many phases of operation, instrumentation, maintenance, etc., must be performed by remote control, as indicated in Chapter VII in connection with chemical processing plants. A great deal of work has been, and is being, done on the development of remote-control devices for the handling of radioactive material both in the laboratory and in the plant. The situation may become particularly acute in connection with the operation of power reactors; for it may then be necessary to discharge and recharge the fuel without shutting the reactor down.

9.80. Remote-control operation, in general, involves the use of established mechanical and electrical principles, but the problems are somewhat novel in character, and, consequently, they represent a challenge to the engineer whose interest lies in this direction. At the Argonne National Laboratory a Remote Control Engineering Division was established in 1947,† and similar groups exist at other A.E.C. contractor installations.‡

Safety Measures

9.81. Shielding for a reactor and related equipment is constructed so that operating personnel normally receive considerably less than the maximum radiation dose of 60 mrem/working day, i.e., 300 mrem/5-day week. Nevertheless, there is a possibility of leakage, especially of neutrons, through cracks or other openings, particularly if the shield has removable sections. A thorough survey of the entire shield should thus be undertaken as a reactor is brought up to full power for the first time. If radiation leaks are found, steps should be taken to repair them or to restrict access to the danger area.

* K. Z. Morgan, *Chem. Eng. News*, **25**, 3794 (1947); *Eng. J. (Canada)*, **31**, 154 (1948); "Medical Physics," Yearbook Publishers, Inc., 1950, Vol. II, p. 766; G. W. Morgan, *Nucleonics*, **4**, No. 3, 24 (1949); J. M. Smith, Jr., *ibid.*, **11**, No. 6, 41 (1953).

† *Nucleonics*, **10**, No. 11, 33–51 (1952).

‡ See series of papers in *Proc. Conf. on Nucl. Eng.*, Berkeley, 1953, Section G.

9.82. It is very improbable that personnel connected with reactor and associated operations will receive an excessive dose from the radiation that penetrates the shielding. However, there are various maintenance activities, both routine and emergency, which may require the handling of radioactive materials.* In such cases some protection may perhaps be attainable by the use of temporary or mobile shielding or, if possible, by performing the operation at a distance with the aid of suitable tools. All work in a hazardous area must be carried out in cooperation with a radiation surveyor who will determine, from the observed dosage rate, the maximum permissible exposure time.

9.83. Access to and egress from an area where there is a possibility of radioactive contamination being present should be by definite routes to prevent spread of the contamination. Personnel entering the area are supplied with expendable clothing, gloves, and shoe covers; these are left at the prescribed exit for decontamination or disposal, according to circumstances. Before being dismissed, each worker is subjected to careful monitoring (§ 9.119).

9.84. Special precautions must be taken to prevent radioactive material from entering the body. Strict personal hygiene must be practiced in areas where there may be contamination, and there should be no smoking, eating, or drinking in areas where such actions might result in an appreciable intake of radioactive material. The ventilating system for reactor and other buildings should be designed so that the air always flows from regions of lower radiation level to those of higher level. Dust must be controlled, e.g., by vacuum cleaning if possible; if sweeping is required, there should be enough moisture present to prevent dust being raised. In areas where some dust in the air is unavoidable, operating personnel should wear suitable masks.

Monitoring: Air and Water Surveys

9.85. The monitoring operations performed in order to protect personnel from receiving excessive doses of radiation may be divided into three broad classes: (1) general monitoring of air, water, and areas; (2) more detailed survey of working areas and of equipment; and (3) monitoring of personnel, clothing, etc. Although the various functions may be distributed differently in different A.E.C. installations, the foregoing classification is convenient for purposes of description. It should be mentioned, too, that the measurement techniques described below may vary from one place to another and that, in any event, the whole field of radiation instrumentation is always undergoing change.

* See C. B. Wagner, *Elec. Eng.*, **74**, 221 (1955).

9.86. An important hazard in air consists of dust particles that have become radioactive, and several different instruments are used for removing the particles and determining their activity.* For continuous automatic monitoring of the atmosphere in areas within or surrounding a reactor site, the *constant air monitor* can be used; this is a device for measuring the particulate beta and gamma activity in the ambient air. A vacuum pump draws air continuously through a filter paper wrapped around a Geiger-Müller (G-M) tube (see § 5.78) mounted in a lead shield. The tube is connected with a count-rate meter which is attached to an automatic recorder. A continuous record is thus obtained of the rate of collection of activity and of the total activity collected in a given time. An associated relay-alarm system provides audible warning when a predetermined radiation level is reached. The filter paper upon which the dust particles collect is changed at regular intervals, usually daily.

9.87. Two other instruments are used to collect the dust present in a known volume of air; the activity of the material collected is then determined in a counting laboratory. The *Filtron* is a semiportable instrument consisting of an electric motor driving a blower unit which forces air at the rate of about 5 ft^3/min through a chamber containing a filter paper. The collection period is generally about 30 min. About 70 per cent of all particles larger than 0.5 micron (1 micron = 10^{-4} cm) entering the chamber remain on the filter paper. The instrument is used to determine beta and gamma activities; it is not recommended for alpha-particle determinations because of spurious counts given by the fiber of the filter. In the *Precipitron* the blower forces the air through a corona discharge produced by a high voltage, so that the dust particles acquire electrical charges. They are then precipitated electrostatically on to a thin aluminum foil. The instrument has a collection efficiency of 90 per cent or more for particles between 0.1 and 10 microns.

9.88. For checking on the background activity of various zones around a reactor or other source of radiation, gamma-monitoring systems of a permanently installed type are desirable. The dosage rate in various locations can then be recorded at a central point. Thus, not only are personnel entering the area informed in advance of the existing radiation levels, but significant increases can be detected and steps taken to determine the cause and to correct the situation.

9.89. An instrument known as a *Monitron* is often used to record the background radiation in working areas. It consists of a large ionization chamber which is coated with carbon, for the detection of gamma radiation only, or with enriched boron (§ 5.33) if it is to be used for thermal neutrons in addition to gamma rays. The chamber is connected with an amplifier circuit and a count-rate meter which is calibrated to read directly in mr/hr. A relay system, actuating an alarm, may be set to provide a warning signal at any desired gamma-

* For the importance of air monitoring see M. Eisenbud, *et al.*, *Nucleonics*, **12**, No. 8, 12 (1954).

radiation level. Actual thermal-neutron intensities are not measured, but relative values are indicated.

9.90. If water is used as either primary or secondary coolant for a reactor, it is generally discharged ultimately to a public stream. Monitoring is therefore necessary to avoid the direct hazard that might arise from drinking the water or the indirect hazard resulting from the accumulation of radioactivity in various forms of aquatic life. When water is the primary coolant, as at Hanford, it leaves the reactor with a certain amount of induced activity due to the capture of neutrons by traces of dissolved salts. The water is held for several hours in order to allow the short-lived activity to decay, but it is always monitored before being returned to the Columbia River.

9.91. The ideal method for monitoring water would be to use an immersion or dipping type of G-M tube, whereby a continuous record of the activity of the effluent could be obtained. Although such instruments exist, the procedure is not too satisfactory because the tube becomes contaminated with continued use. One way of overcoming this difficulty is to take measurements periodically, thus greatly reducing the time of contact of the tube with radioactive contaminants. An alternative is to place a sensitive G-M tube just above the surface of a large volume of water, e.g., a retention basin. In any case it is desirable that samples of water be taken to the laboratory regularly, where they are evaporated to dryness and the residues counted or subjected to radioanalysis. Consideration must be given to the possible loss of volatile elements during the evaporation.

9.92. In addition to cooling water, it is frequently necessary to discharge dilute aqueous solutions containing traces of radioactive material as an aspect of waste disposal. These solutions are usually held up in tanks until the activity has largely decayed, and then they are gradually discharged into settling basins from which they find their way into the surrounding soil and possibly into an existing drainage system. As a protective measure, samples of water and soil from various locations are regularly taken to the laboratory for appropriate analysis.

Radiation Survey Instruments*

9.93. All exposed surfaces, especially working surfaces, and equipment which has to be approached or handled must be subjected to a detailed radiation survey. In addition, operations involving radioactive material should be monitored so as to make sure that persons performing the operation do not receive an excessive dose of radiation. The instruments used must be portable and reasonably robust, and they should respond rapidly, either visually or audibly. The type of instrument chosen for a particular radiation survey depends on (1) the nature of the radiation, e.g., alpha, beta, or gamma radiation or neutrons; (2) the magnitude of the dosage rate; and (3) whether qualitative (detection) or

* See Nuclear Instrument Handbook, *Nucleonics*, **4**, No. 5, 97–152 (1949).

quantitative (measurement) information is required. It may be noted that it is a common practice to make qualitative observations first and to follow them with a quantitative survey if required. Information concerning the type of radiation is generally available from the nature of the operation and of the materials involved. If this is not the case, a preliminary scanning survey may be necessary in order to determine which instrument is most suited to the circumstances.

9.94. Many instruments, made by different manufacturers, are available for radiation surveys, but most of them fall into a few general categories determined by the purpose for which they are intended. The descriptions given here may be taken as fairly typical, although there may be variations due either to personal preferences or to improvements in design which are continually being introduced. For purposes of clarity, the treatment will be divided into two parts; the first will deal with survey instruments used for detection purposes and which are not intended to provide a reliable quantitative indication of the radiation dosage, and the second will be concerned with devices which are used for actual quantitative measurements of the various radiations.

Detecting Instruments

9.95. For the detection of beta and gamma radiations some form of *G-M survey meter* is generally used. It is employed in decontamination operations and for



Fig. 9.3. G-M Survey Meter (Berkeley Division, Beckman Instruments, Inc.)

general survey of personnel, clothing, protective equipment, tools, etc., where a qualitative indication of the activity, often at a relatively low level, is all that is required. The G-M tube is contained in a probe attached to the portable instrument box by means of a cable about 36 in. long (Fig. 9.3). The high voltage required to operate the tube is supplied by batteries or by a vibrator power supply contained in the box.

9.96. The pulses from the tube, arising from the presence of radiation, are amplified and fed to a pair of headphones for aural detection. The instrument is also equipped with a count-rate meter calibrated roughly in mr/hr, from 0.05 to 20 mr/hr, but the readings are not too accurate, except at the energy for which the system was calibrated. Normally, the G-M survey meter detects gamma radiations and beta particles of energy 0.2 Mev or more, but by the use of a shield it is possible to determine whether the activity detected is due to beta particles or gamma radiations or both. Because of its great sensitivity, the instrument is particularly suited to the detection of low-level activities, but it should not be used for quantitative measurement of dose rates.

9.97. A portable instrument for the detection of alpha particles is the *alpha-survey meter;* this is a battery-operated proportional-counter type of instrument with headphones for aural detection. The detecting chamber may be included in the instrument box, or it may be contained in a separate probe. In some forms a count-rate meter, capable of indicating up to 80,000 alpha particles/min, semiquantitatively, is included. By adjustment of the sensitivity of the electronic circuit, the relatively small pulses produced by beta particles and gamma rays are not counted, and so the instrument responds only to alpha particles.

9.98. When the alpha-particle survey is carried out at certain fixed locations, e.g., when checking personnel for contamination, an a-c-operated proportional counter, known as *Poppy*, is sometimes used. It has a separate probe, and audible indication of the presence of alpha particles is provided by a loud speaker. Poppy is designed for the detection and semiquantitative measurement of low-level alpha radiation greater than 1000 particles/min. The instrument is insensitive to gamma radiation below 1 r/hr and to moderately high levels of beta activity, so that it detects essentially only alpha particles. Portable forms of the Poppy instrument have been constructed, with the operating voltage supplied by batteries, but they have not proved too satisfactory.

9.99. The detection of slow neutrons can be performed with a proportional-counter survey meter, similar to that used for alpha particles, containing boron trifluoride, preferably enriched in boron-10. If the walls are not too thin (so that alpha particles cannot penetrate them) and the counting circuit is adjusted so that it is not sensitive to beta and gamma radiation, the instrument will respond only to slow neutrons.

9.100. For detecting fast neutrons a portable instrument, called a *fast-neutron dosimeter*, operating in the proportional region, is used; it responds to the large pulses caused by recoil protons produced by passage of fast neutrons through

a thin layer of paraffin. Neutrons in the energy range from 0.1 to 10 Mev are detected. The electronic circuit is designed so that the meter does not respond to small pulses and it is consequently insensitive to gamma radiations at dose rates less than 1 r/hr. The same instrument is used both for detection and measurement.

Measuring Instruments

9.101. One of the most accurate devices for the measurement of gamma radiation, regarded as a basic instrument in quantitative monitoring, is the *Lauritsen electroscope*. In addition to being both accurate and sensitive, it is portable and rugged. It is actually an integrating ionization chamber of the electrostatic type, referred to in § 5.25. One of the electrodes is a very fine, gold-plated

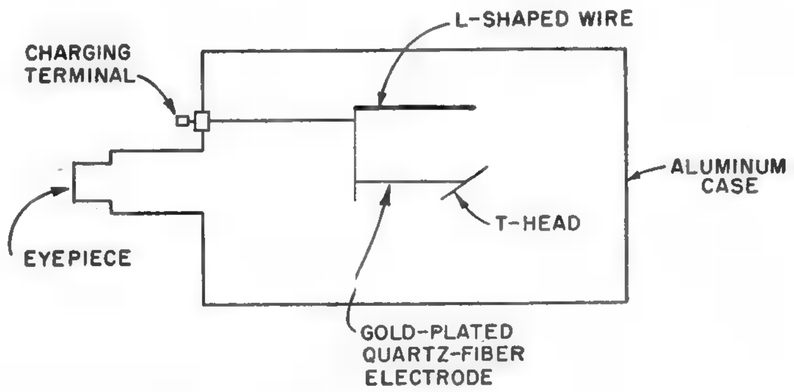


Fig. 9.4. Lauritsen electroscope

quartz fiber connected to a rigid L-shaped metal wire and mounted within, but insulated from, a cylindrical aluminum case. The latter acts as the outer boundary of the air-filled ionization chamber and also as the second electrode (Fig. 9.4). The electrodes are charged to a potential difference of about 200 volts by means of a self-contained battery. The repulsion between the quartz fiber and the L-shaped wire causes the former to be displaced from its normal (discharged) position. If the instrument is now exposed to ionizing radiation, the charge on the fiber decreases and it tends to return to the discharged position. The rate of motion of the fiber, determined by the time taken for its image to move between two fixed points on a scale in the eyepiece of a microscope, is proportional to the radiation dose rate. The Lauritsen electroscope is accurate to within 1 per cent over the range from 1 to 1000 mr/hr of gamma radiation. Although used mainly for the measurement of gamma-radiation dose rates, the instrument, with suitable "windows," can be adapted to the detection of alpha and beta particles.

9.102. A disadvantage of the Lauritsen electroscope is that it does not indicate dose rates directly, since it requires observation over an interval of time.

Consequently, for many quantitative surveys of gamma radiation, another instrument, called the *Cutie Pie* (or C.P.) meter, is frequently used. Although it is not very accurate (±10 per cent), it is fairly robust, is light in weight, and can be carried and read very easily (Fig. 9.5). The C.P. consists of an air-filled ionization chamber made of a Bakelite, which is approximately air equivalent. The dose rate, proportional to the ion current, is indicated directly in mr/hr on a familiar type of pointer microammeter. The meter has three ranges, which may differ somewhat in instruments made by different manufacturers; the overall range is usually from 5 to 5000 or 10,000 mr/hr. For the measurement of gamma rays of low energy, or when it is required to detect beta particles in addition, a thin-walled chamber of special (air-equivalent) black paper, supported

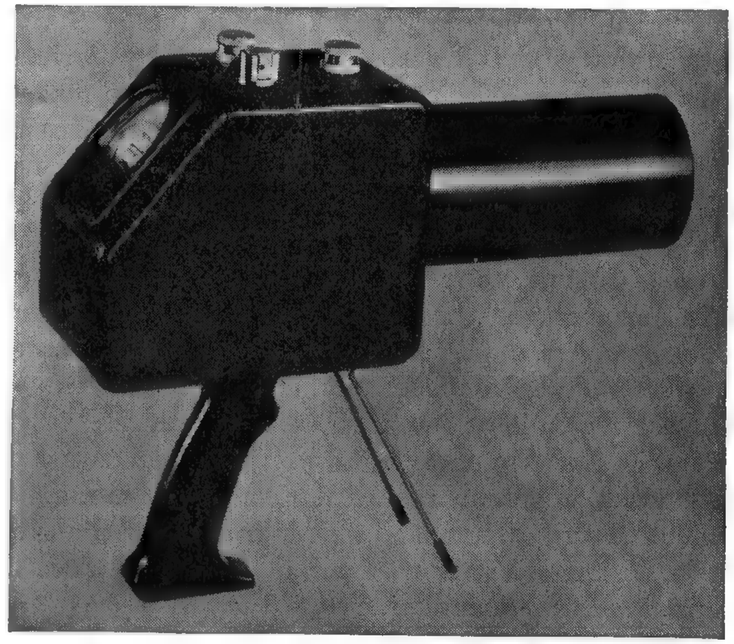


Fig. 9.5. "Cutie Pie" meter (Tracerlab, Inc.)

by a Bakelite frame, is used. With the aid of a very thin "window," alpha particles of fairly high energy can also be detected. If the interior of the chamber is coated with enriched boron, the C.P. will measure slow-neutron flux.

9.103. A modification of the C.P. meter, called the *Fish-Pole Probe*, is used for high levels of radiation, e.g., 1 to 100 r/hr, to an accuracy within about 10 per cent. Depending upon the type of ion chamber and the sensitivity of the associated meter, relative levels of beta and gamma radiation up to several hundred r/hr can be indicated, but not measured with any degree of accuracy. The Fish-Pole Probe consists of an ionization chamber attached to the end of a pole which can be extended to about 12 ft. Thus, the observer, holding the other end of the pole, can stay at a distance from the source where the radiation level

is relatively low. The electrical circuit is similar to that in the C.P. instrument, and the dose rate is read in the same manner, by means of a microammeter attached to the observer's end of the pole.

9.104. For the detection and measurement of very low levels of gamma radiation (and X-rays) a portable scintillation counter has been found very useful, because, as stated in § 5.87, instruments of this type have a high efficiency. The general range of the scintillation counter employed in this connection is from 0.005 to 5 mr/hr. It is designed so as to be insensitive to alpha and beta particles.

9.105. The alpha-survey meter described in § 9.97 is also used for the semiquantitative measurement of contamination of benches, desks, floors, etc., by alpha-particle emitters. It has a range of from about 300 to 80,000 particles/min, for alpha particles having energies of 2 Mev or more. An external shield is used to determine whether radiations other than alpha particles are present.

9.106. For the measurement of the low thermal-neutron fluxes which might be permissible in an operating area, the most accurate device is a Lauritsen electroscope lined with enriched boron. If there is appreciable gamma radiation, two similar instruments should be used; one of these is lined with boron and measures both gamma radiation and slow neutrons, whereas the other, which is not so lined, measures only the gamma radiation. The difference in the readings when the two chambers are in the same location is consequently a measure of the slow-neutron dose rate. The combination described can determine slow-neutron fluxes up to about twice the maximum permissible average value of 2000 neutrons/(cm²)(sec) for a 40-hr week. For higher fluxes, very approximate measurements can be made by means of a proportional counter containing enriched boron trifluoride in conjunction with a count-rate meter.

9.107. Fast neutrons are determined quantitatively by the same instrument (the fast-neutron dosimeter) as is used for detection purposes (§ 9.100). Its response is proportional to human-tissue sensitivity over the energy range from 0.2 to 10 Mev, and fast-neutron fluxes from 10 to 10,000 neutrons/(cm²)(sec) can be measured by means of a count-rate meter.

9.108. The information contained in this section concerning instruments used for survey purposes is summarized in Table 9.9. All the devices, except the a-c-operated Poppy, weigh about 10 lb or less, so that they may be regarded as readily portable; the weight of the C.P. meter is only 4 lb.

Personnel Monitoring

9.109. All persons working within areas where nuclear radiations may be encountered must wear individual personnel monitoring devices; two types of such instruments are in general use: (1) pocket (ionization chamber) meters and (2) film badges (or film dosimeters). In addition to these meters which record the total radiation dose received over a period of time, individuals leaving a contaminated area are surveyed by appropriate counters which will be described later.

TABLE 9.9. INSTRUMENTS USED IN RADIATION MONITORING

Instrument	Operating Region	Radiation	Approximate Useful Range	Remarks
Detecting Instruments				
G-M survey meter	G-M region	Gamma and beta (>0.2 Mev)	0.05 to 20 mr/hr	Not quantitative
Alpha-survey meter	Proportional counter	Alpha	300 to 80,000 dis/min	Semiquantitative
Poppy (a-c-operated)	Proportional counter	Alpha (low levels)	Low levels >1000 dis/min	Semiquantitative
Fast-neutron dosimeter	Proportional counter (with paraffin)	Fast neutrons	10 to 10,000 n/(cm²)(sec)	Quantitative (see below)
Slow-neutron survey meter	Proportional counter (with BF_3)	Slow neutrons	Relatively high thermal fluxes	Semiquantitative at high fluxes
Measuring Instruments				
Lauritsen electroscope	Ion chamber (electrostatic)	Gamma (can detect alpha and beta)	1 to 1000 mr/hr	Accurate to ±1%
C.P. (Cutie Pie)	Ion chamber (current indicating)	Gamma (can detect beta)	5 to 10,000 mr/hr	Light and robust; accurate to ±10%
Fish-Pole Probe	Ion chamber (current indicating)	Gamma (high level)	1 to 100 r/hr	C.P. meter on 12-ft pole
Scintillation counter	Scintillation	Gamma (low level)	0.005 to 5 mr/hr	Very efficient and sensitive
Fast-neutron dosimeter	Proportional counter (with paraffin)	Fast neutrons	10 to 10,000 n/(cm²)(sec)	.....
Lauritsen electroscope boron lined	Ion chamber (electrostatic)	Slow neutrons (low level)	Up to 4000 n/(cm²)(sec)	Requires compensation if gamma present
Slow-neutron survey meter	Proportional counter (with BF_3)	Slow neutrons (high level)	Relatively high thermal fluxes	Semiquantitative

Pocket Meters

9.110. The pocket meter (or chamber) is actually an ionization chamber, similar to a fountain pen in size and shape. It consists of a cylinder of plastic material lined internally with graphite which acts as the positive electrode. A stout wire, supported at its ends by insulators, runs axially through the cylinder and serves as the negative electrode (Fig. 9.6). The electrodes are charged in a charger-reader (or *minometer*) to a known potential difference, about 150 volts (Fig. 9.7). If the insulation is satisfactory and there is no exposure to radiation, the charge will remain unchanged. If ionizing radiations enter the chamber there is a decrease of potential which is proportional to the amount (or dose) of radiation received. The change in potential is measured, at the end of the day, by means of an electrometer which is part of the minometer, and the corresponding radiation dose is recorded.

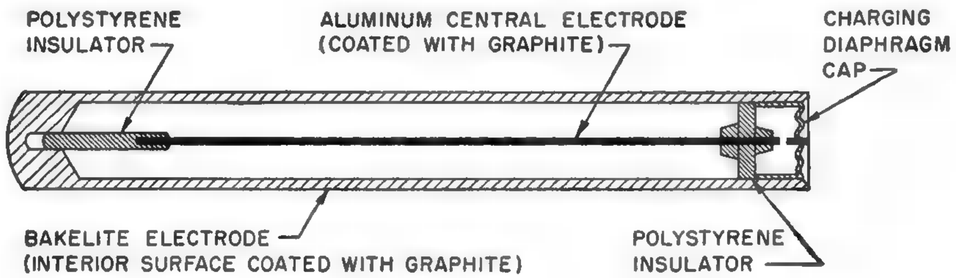


Fig. 9.6. Pocket ionization chamber

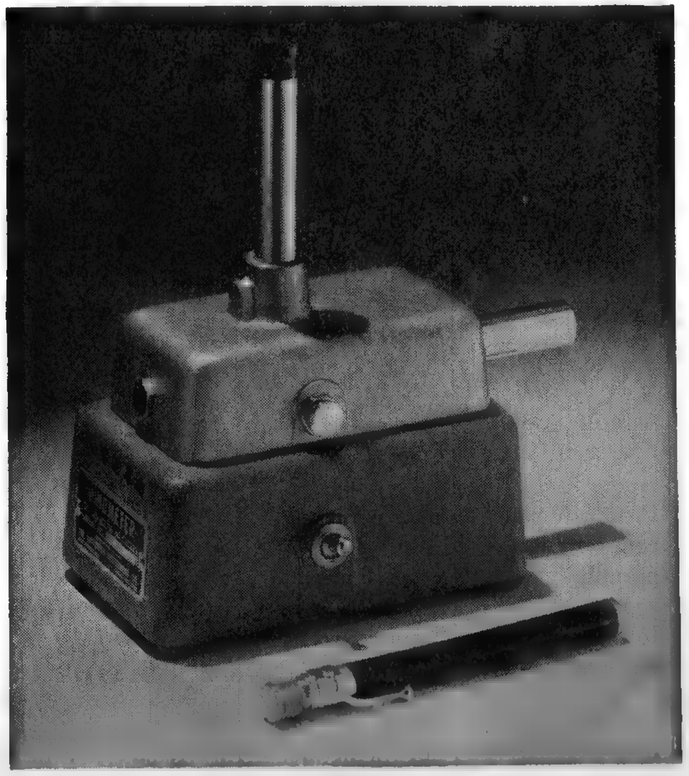


Fig. 9.7. Charger-reader for pocket chambers (Victoreen Instrument Co.)

9.111. As a general rule, each individual carries two pocket chambers. The one indicating the lower exposure is taken as giving the correct dosage, since various electrical and mechanical factors tend to make the readings too high. Pocket meters measure gamma radiations and such beta particles (energy about 1 Mev or more) as are able to penetrate the walls of the chamber. Their range is usually up to 200 mr total dosage and the accuracy is within 10 per cent. Pocket chambers have been made with boron linings, so that they are also sensitive to slow neutrons.

9.112. Where large radiation exposures are possible in the course of an operation, it is desirable that each individual should be able to keep track of the total dose received from time to time. For this purpose self-reading pocket dosimeters are used. These are also similar to a fountain pen in appearance and operate in the ionization chamber region. The outer electrode is cylindrical and the inner electrode consists of a short wire to which is attached a flexible, metal-coated quartz fiber, similar to that in the Lauritsen electroscope (Fig. 9.8). When the dosimeter is charged, the quartz fiber diverges from the wire, but when radiation enters the chamber the deflection gradually decreases. The position of the quartz fiber on a calibrated scale, as observed through a lens system fitted into one end of the instrument, gives the total radiation dose received up to the time of reading. The self-reading dosimeter usually indicates exposures up to 200 mr, although instruments with higher ranges are available.

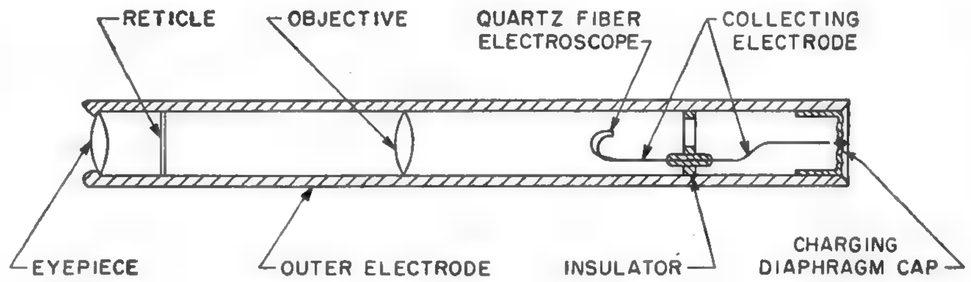


Fig. 9.8. Self-reading pocket dosimeter

Film Dosimeters

9.113. The action of the film dosimeter is dependent on the fact that ionizing radiations produce an effect on photographic film similar to that of light; upon development, after exposure to radiation, a general blackening is observed. The extent (or density) of the blackening is a measure of the total amount of radiation to which the film has been exposed, although there is some energy depend-

ence as will be indicated later. Highly ionizing particles, such as alpha particles and protons, form definite linear tracks in their motion through the photographic emulsion. If a special fine-grain (nuclear track) film is employed, the tracks are particularly clear after development and so they can be readily seen (and counted) by means of a microscope. This type of film is used for the determination of neutron exposure. Fast neutrons produce tracks due to recoil protons, whereas slow neutrons liberate protons as a result of the $N^{14}(n, p)C^{14}$ reaction with nitrogen in the film.

9.114. The film-badge dosimeter consists of a small frame of metal or plastic containing one or two packs of film, about $1\frac{1}{2}$ by 2 in. in size (Fig. 9.9); one pack contains beta- and gamma-sensitive film and the other holds nuclear-track film for the determination of neutron exposure, if this is required. For beta- and

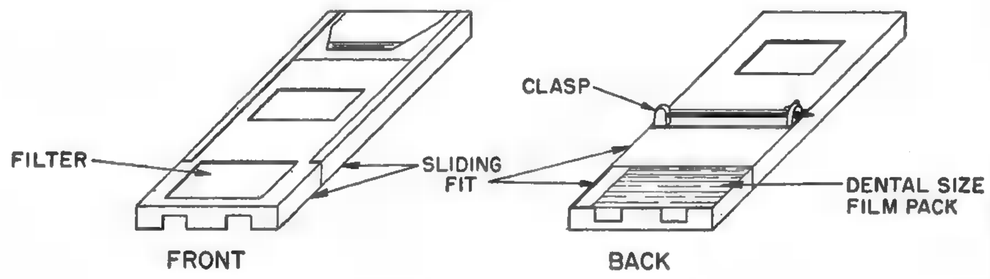


FIG. 9.9. Film-badge dosimeter

gamma-radiation measurement two or three films of different sensitivity are in the pack, and these may cover a total dosage range from 50 mr to 20 r or to as much as 500 r* when exposure is to gamma radiations of energies from 0.3 to 2 Mev. A single film is generally adequate for neutron determination, and this may cover neutron exposures from the maximum permissible level to a hundred times this value.†

9.115. In the assembly of the film dosimeter, each of the film packs, wrapped in paper to prevent access of light, is placed in the frame, with the neutron-sensitive film at the back. In front, and covering part, of the film packs are one or more thin metallic shields (about 1 mm thick), one of which consists of cadmium. The cadmium shield serves several purposes. In the first place it helps to make the film respond uniformly to gamma radiations of all energies. It happens that the sensitivity of photographic film to gamma radiation increases very markedly for energies less than about 0.3 Mev; thus 1 r of radiation of 0.1 Mev energy will produce six or more times the blackening caused by the same dose at 1 Mev. However, passage through a shield of a heavy metal results in the absorption of a considerable proportion of the low-energy radiation (cf. Fig. 2.11), and so the response of the film is more uniform over a range of energies.

* Measurement in the high range may prove useful in the event of a major accident.

† For film monitoring of fast neutrons, see J. S. Cheka, *Nucleonics*, 12, No. 6, 40 (1954).

The second purpose of the metal shield is to absorb beta particles; hence the beta-gamma film under the shield is affected only by gamma radiations, whereas the unshielded part responds to both gamma rays and to beta particles which pass through the paper enclosing the film, i.e., with energies in excess of 0.1 Mev. Finally, since cadmium is such a good absorber of slow neutrons, it is penetrated only by fast neutrons. The shielded portion of the neutron-sensitive film will thus indicate fast neutrons, but the unshielded part will be exposed to both fast and slow neutrons.

9.116. At certain prescribed intervals, usually at the end of each week, the film dosimeters are taken to the laboratory to be developed. The amount of blackening of the beta-gamma films, as determined by means of an electronic densitometer, gives the total exposure to both beta and gamma radiations and to the latter alone. These are indicated by the densities of the unshielded and shielded parts of the film, respectively. The films are calibrated in terms of roentgens by means of a radium standard (cf. § 9.38). The developed films are filed to provide a more or less permanent record of the radiation exposure of the individual.

9.117. Neutron-sensitive films, after development, are examined with a microscope, and the tracks produced by fast and slow neutrons, under the unshielded portion, and by fast neutrons alone, under the cadmium-shielded portion, are counted. Weekly inspection of neutron films is necessary only for those persons who are likely to be exposed to such radiations to an appreciable extent. For others, examination at intervals up to three months or more may be sufficient.

9.118. It was mentioned in § 9.115 that in some film dosimeters other metallic shields, in addition to cadmium, are used in front of the beta-gamma film; each of these shields then covers a separate part of the film. The purpose of such shields is to provide information concerning the energy of the gamma radiation. The absorption coefficients of different metals, e.g., cadmium, copper, and lead, vary differently with energy, and by comparing the densities of the blackening produced under shields of these several metals some conclusions can be drawn concerning the energy of the incident gamma radiation.

PERSONNEL SURVEY INSTRUMENTS

9.119. In addition to the meters described above, which give the total accumulated radiation dose over a period of time, an aspect of personnel monitoring is the radiation survey of individuals leaving an area where contamination is possible. One method employed is to use a G-M survey meter and to run the probe containing the tube over all parts of the body to detect the possible presence of radioactivity. If the individual or his clothing is found to be contaminated, appropriate decontamination measures must be taken.

9.120. A stationary (nonportable) device for personnel surveying, generally located at a point of egress from an area where contamination may be encountered, is the *hand-and-foot counter*. This consists of an arrangement of five

separate G-M tubes and their associated count-rate meters capable of detecting essentially all gamma radiations and beta particles with energy in excess of 0.2 Mev, but not alpha particles. The individual steps on to a platform and places his hands in two slots provided for the purpose. The counting mechanism is started by pressing down with the hands, and it stops automatically after 24 sec. The five indicators then register the respective amounts of contamination on the palm and back of each hand and on both shoes. If the amounts exceed certain tolerance limits, the individual must report for decontamination.

RADIOACTIVE WASTE DISPOSAL*

General Considerations

9.121. An aspect of radiation protection, which is important for persons not connected with reactor operation, as well as for those directly associated with it, is concerned with the disposal of radioactive wastes. In this connection the term "waste" is employed in a general sense to refer to any material, including air, which must be discharged to the surroundings. The disposal of this material, much of which is dangerously radioactive, must be carried out in such a manner as neither to constitute a hazard to human beings nor to interfere with the existing ecological equilibrium. The most significant source of radioactive wastes is the chemical processing of spent reactor fuel, and the disposal of these wastes was considered briefly in Chapter VII. However, even the reactor itself produces wastes, such as water and air used as coolants, ventilation air, contaminated equipment, liquid from cleaning and decontamination operations, etc. In a reactor using liquid fuel, gaseous fission products such as krypton, xenon, and iodine will be liberated and must be disposed of. Further, many reactor installations will have associated laboratories from which a variety of radioactive wastes will arise.

9.122. The problem of waste disposal is greatly complicated both by the variety and the variability of the material to be treated from day to day. At Oak Ridge National Laboratory, liquid wastes are divided into three categories in order to simplify, to some extent, their treatment. These are (1) *metal wastes*, containing dissolved compounds of uranium or other elements which must be recovered; (2) *radiochemical wastes*, consisting chiefly of highly radioactive fission products and various by-products from development and other processes; and (3) *process wastes*, which are large volumes of liquid, mainly cooling water and nonradioactive or feebly radioactive process by-products. In view of the complexity of the situation, it should be one of the prime considerations in the

* Report TID-5031, pp. 353, 400; F. Western, *Nucleonics*, **3**, No. 2, 43 (1948); Symposium on Radioactive Wastes, *Ind. Eng. Chem.*, **43**, 1499–1544 (1951); "Handling Radioactive Wastes in the Atomic Energy Program," U. S. Government Printing Office, 1951; C. P. Straub, *Nucleonics*, **10**, No. 1, 40 (1952); L. P. Hatch, *Am. Scientist*, **41**, 410 (1953); W. A. Rodger, *Chem. Eng. Prog.*, **50**, 263 (1954); B. Manowitz and L. P. Hatch, *Chem. Eng. Prog. Symposium Series*, No. 12, **50**, 144 (1954). For a bibliography of radioactive waste disposal, see Report TID-375.

design and operation of nuclear reactors and their associated chemical plants and laboratories to keep to a minimum the quantity and variety of radioactive material which has to be treated as waste.

9.123. Apart from the utilization of natural decay to reduce the activity of waste materials, two general methods of controlling radioactive wastes have been used: (1) dilution and dispersion, and (2) concentration and storage. To apply them satisfactorily requires a knowledge of the geology of the area and also of the meteorological conditions. For example, in certain circumstances radioactive material placed in the ground might be perfectly safe, whereas under other geological conditions contamination of the surrounding water supplies might result. Climate is also important because atmospheric inversions, rainfall, and winds might tend to spread contamination over the ground. In connection with the dilution and dispersion of radioactive wastes, either in the atmosphere or into rivers, etc., the A.E.C. places limits upon the activity and amounts of material that may be released. The objective is to prevent the radioactivity in the air and in water supplies from reaching the maximum permissible concentrations given in Table 9.7.

9.124. The question of waste disposal in relation to the surrounding population, geology, and climate should be considered in selecting a site for a reactor or radiochemical plant. A poor choice in this respect could lead to considerable difficulties in operation. Cooperation of sanitary engineers and industrial hygiene engineers with health physicists is desirable, both in the interest of protection of the public as well as that of efficient and continuous functioning of the reactor plant.

9.125. Although the procedures for dealing with radioactive wastes mentioned in Chapter VII and to be described below have proved adequate so far, it should be emphasized that they are essentially of a local and provisional nature. For example, storage of contaminated material by burial requires increasing areas of ground and obviously this cannot continue indefinitely. It is evident, therefore, that if there is to be extensive development of nuclear power, more economical and effective methods for waste disposal will have to be worked out.

Disposal by Dilution and Dispersal

9.126. Radioactive gaseous wastes are usually allowed to mix with large volumes of air in the atmosphere so that they are thereby considerably diluted and dispersed. Thus the cooling air of the Oak Ridge and Brookhaven reactors is disposed of by discharging it to the atmosphere through high stacks.* The off-gases from spent-fuel-element dissolvers are treated in a similar manner (§ 7.209). If the air or other gas contains suspended particles, these particles should be re-

* The chief radioisotopes present in air that has passed through a reactor are oxygen-19 (half life 29 sec), formed by $O^{18}(n, \gamma)O^{19}$; nitrogen-16 (half life 7.3 sec), formed by $O^{16}(n, p)N^{16}$; and argon-41 (half life 1.83 hr), formed by $A^{40}(n, \gamma)A^{41}$. The argon-41 is the most significant of the three.

moved by electrostatic precipitation or by filtration before the gas is discharged. The radioactive solid which is collected is then usually buried (§ 7.214).

9.127. Cooling water and aqueous wastes of relatively low activity may be stored for a time to permit the activity to decrease to some extent.* The liquid is then diluted at a controlled rate with a large volume of water, as in a river, and the activity is thus dispersed The ultimate concentration of radioactive material in the water must not exceed the maximum permissible levels. To insure the absence of hazard, measurements are made regularly on the activity of the river water, in fish, and in other aquatic life.

9.128. Because of the desirability, for reasons given in Chapter VIII, for removal of as much as possible of the dissolved material present in water to be used for reactor cooling, the problem of disposal of the effluent is greatly simplified. As a result the activity induced in the water is much less than would otherwise have been the case. The procedure of prior purification is used in connection with the cooling water for the Hanford reactors. As stated above, after passage through a reactor, the cooling water is held for several hours, and then mixed off with the Columbia River. The low-level aqueous wastes from Oak Ridge National Laboratory are disposed of in a similar manner; after retention for several days, first in a settling basin and then in White Oak Lake, the waste liquid is gradually discharged into the Clinch River. The liquid wastes of low activity from the British (Windscale) plutonium production plants are pumped through pipe lines into the sea at a distance of about two miles from shore.†

Disposal by Concentration and Storage

9.129. Solutions which are too radioactive to be disposed of by dilution and dispersal can be treated by concentration; this decreases the bulk of active material, which is then buried either on land or at sea, or it may be held in underground storage tanks. Several methods of concentration have been proposed and applied in various instances. The most successful procedure employed hitherto is evaporation. If the condensate has little or no activity, it can be disposed of very readily, but there may be circumstances in which this is not the case. For example, the presence of radioelements which are themselves volatile or which form volatile compounds may cause the condensate to be radioactive. In addition there is always the possibility that some active material will be carried over from the waste solution into the condensate by entrainment in the vapor.

9.130. Evaporation has been proved to be a satisfactory method for concentrating active wastes, but it is expensive and so other procedures have been used. Among these, mention may be made of (1) precipitation of the active species with a suitable carrier and (2) removal with an ion-exchange resin. Both of

* The activity of effluent cooling water is due mainly to sodium-24 (half life 15 hr), formed by $Na^{23}(n, \gamma)Na^{24}$, although various other radioisotopes are frequently also present.

† K. E. B. Jay, "Britain's Atomic Factories," H. M. Stationery Office, 1954, p. 38.

these methods can result in greater concentration than is practical by evaporation and, probably, at lower cost, although evaporation gives a higher degree of decontamination. The precipitation process is essentially the same as the flocculation method commonly employed in conventional water treatment. An interesting application of the ion-exchange principle is based on the use of montmorillonite, a clay-like material with a high capacity for exchange with ions of the most important fission products. After the clay has taken up the maximum amount of these ions, it is heated to 1000°C (1830°F), and as a result its structure is changed so that only a very small proportion of the ions can be re-extracted.*

9.131. Another process which has been demonstrated successfully, although it has not been applied on a large scale, makes use of biological assimilation. By utilizing established sewage-disposal techniques, various organisms normally occurring in sewage or in sewage processes have been found to concentrate certain elements. The solution must, of course, provide sufficient nutrient material to promote growth of the organisms.

9.132. The radioactive liquid concentrates obtained by any of the methods described above are stored in various ways. They may be placed in underground storage tanks, or they may be buried directly in the ground. In the latter case it is important that the soil should be relatively nonporous, otherwise some of the activity may seep into the water-supply system. A further possibility is to incorporate the concentrated liquid waste into concrete blocks which are then buried on land or at sea.

9.133. When the soil has very good ion-exchange properties, the concentration and burial stages may, in a sense, be combined by running the waste solution directly into pits. As the liquid percolates through the earth, the radioactive cations are adsorbed and held in the soil by the process of ion exchange. There is, of course, a limit to the amount of active material that can be retained in this manner in any particular location.

9.134. Many solid wastes are already in concentrated form so that they can be stored or buried in a suitable place, preferably in dry or impervious ground. In some instances, however, a decrease in volume can be achieved by compression. If combustible matter is present, e.g., contaminated clothing or wood, it may perhaps be burnt off, but great care must be taken to prevent the spread of radioactive smoke. Considerable work has been done on the development of incinerators for the treatment of combustible radioactive wastes.

9.135. In order to ensure that waste-disposal procedures are properly achieving their objective, various radiation-survey operations, some of which were mentioned earlier, are performed regularly. Samples of water, mud, soil, etc., are taken to the laboratory from time to time for detailed radiochemical analyses. In addition, continuing studies are made of fish and plant life to determine to what extent, if any, they are affected by the discharge of radioactive wastes into their environment. So far, the methods and precautions used have proved com-

* W. S. Glnell, J. J. Martin, and L. P. Hatch, *Nucleonics*, 12, No. 12, 14 (1954).

pletely satisfactory, and there is no evidence that reactor operations have had any appreciable effect on ecological conditions. Nevertheless, there is considerable scope for improvement in the processes for radioactive waste disposal.

Symbols Used in Chapter IX

A	atomic weight (or mass number)
C	number of curies
d	distance from source in cm
E	energy
f	distance from source in ft
g	mass of radioactive material in grams
I	intensity of radiation
I_0	initial intensity of radiation
K	constant
m	milligrams (of radium)
mr	milliroentgen
N	number of atoms per cm^3 (or per gram)
P	reactor power in kw
r	roentgen
rhm	roentgens per hr at 1 meter distance
x	thickness of absorber in cm
γ	fraction of disintegrations accompanied by gamma rays
μ	linear absorption coefficient
μ_e	energy-absorption coefficient
σ	scattering cross section for fast neutrons
ϕ	flux (photons or neutrons/$(cm^2)(sec)$)

PROBLEMS

1. If t_b is the biological half life of an element in a certain organ and $t_\frac{1}{2}$ is the radioactive half life of a particular isotope, derive an expression for the effective half life of that isotope in the specified organ. Show that if either t_b or $t_\frac{1}{2}$ is very short compared to the other, then the effective half life is approximately equal to the shorter of the two. Explain the physical significance of the result.

2. The human body consists of 15 per cent by weight of carbon and 1.0 per cent of potassium. Owing to the presence of carbon-14, there is an emission of 12.5 beta particles/min per gram of carbon in the living organism. Further, all forms of potassium contain 0.011 per cent of potassium-40, a beta emitter with a half life of 1.5×10^9 yr. Calculate the number of microcuries of (*a*) carbon-14 and (*b*) potassium-40 present in a human being weighing 70 kg.

3. A standard radium source of 0.010 curie strength, contained in a platinum capsule of 1.00 mm wall thickness, was located at 35 cm in air from a meter which then indicated a dose rate of 76 mr/hr. What is the error of the meter at this reading?

4. After drawing air for 30 min, at the rate of 5 ft^3/min, through a Filtron, the collected sample was found to have a beta-gamma activity of 4850 counts/min, attributed to iodine-131. The collection efficiency of the Filtron is about 70 per cent and the

counter efficiency ("geometry") is 10 per cent. The normal background is 120 counts/min. Is the air safe for continued use?

5. Iodine-131 (half life 8.0 days) emits a 0.36-Mev photon in 85 per cent of its decays and a 0.64-Mev photon in the other 15 per cent. A 10-curie source of this radioiodine is to be kept under water, with no other shielding. What depth of water would be necessary to insure that no individual will receive a dose greater than one tenth of the maximum permissible exposure during a 40-hr working week?

6. Radiolanthanum-140 (half life 40 hr) emits a number of gamma-ray photons, in addition to beta particles; the most important gamma-ray energies are 1.6 Mev in 77 per cent of the decays, 0.9 Mev in 10 per cent, and 2.5 Mev in 6 per cent. Design a lead carrier for a 5-curie source of radiolanthanum, such that the escaping radiations do not represent any appreciable hazard.

7. An individual spends a total of 24 hr per week in the vicinity of a reactor where the maximum average fluxes are 350 2-Mev photons, 120 thermal neutrons, and 5 fast neutrons/$(cm^2)(sec)$. What is the total dose received in rems?

8. An operator receives a total dose of 0.015 rad of fast neutrons during the course of a 40-hr working week. What would be the maximum flux of 1-Mev gamma radiation to which he might be permitted to be exposed at the same time?

*Chapter X**

SHIELDING OF NUCLEAR REACTOR SYSTEMS†

REACTOR SHIELDING PROBLEMS

General Aspects of Shield Design

10.1. In the design of a nuclear reactor and its associated equipment, provision must be made for the attenuation of escaping nuclear radiations by some form of shielding. Not only is such shielding necessary for the protection of personnel, as explained in the preceding chapter, but a relatively high radiation background will interfere with the satisfactory functioning of instruments used in various aspects of reactor operations and control.

10.2. The character of the shield suitable for a particular reactor is very largely dependent on the purposes of the reactor, so that shield design is to be regarded as an integral aspect of reactor design. A reactor of very low or zero power, such as might be used for critical experiments, would require little or no shielding. In a case of this kind the attenuating effect of distance might be sufficient to reduce the radiation dosage to a value below the maximum permissible exposure (see Chapter IX). On the other hand, a reactor operating at high power would require a shield of considerable thickness. For a stationary system a partial solution may be found in building the reactor underground.

10.3. Another aspect of the relationship between reactor and shield design is apparent in connection with various research reactors. In the Bulk Shielding Facility (Swimming Pool) reactor, used for experimental purposes at Oak Ridge National Laboratory (§ 10.124), the shield consists essentially of 17 ft of water in a concrete tank.‡ The design of the reactor is such that it can be charged, discharged, and controlled from above. Apparatus suitable for experiments can be lowered from the top, so that a direct approach to the sides of the reactor is not necessary. In this case water acts as moderator, reflector, coolant, and shield. When air is used as coolant, so that air ducts must be introduced into

* Material contributed by E. P. Blizard and S. Glasstone.
Reviewed by E. P. Blizard and A. Simon.

† For bibliography, see C. J. Engberg, Report TID-3032; a compilation of data useful in shielding calculations is given by J. Moteff, Report APEX-176.

‡ See § 13.70 for description of a similar reactor.

the reactor, or when access to the interior is necessary for experimental purposes, as in the natural uranium-graphite reactors at Oak Ridge (X-10) and Brookhaven, the use of a water shield is not always practical or convenient.

10.4. For mobile reactors, e.g., for the propulsion of an aircraft, the weight of the shield is of overriding importance; if the shielding is too heavy, the reactor may prove to be unsatisfactory for its intended purpose. In this instance the cost of the shield may be of secondary significance. But for a stationary reactor, such as would be used in a power station, cost becomes the primary factor, and weight is now secondary.

10.5. Wherever possible advantage is taken in shield design of the attenuating effect of distance, according to the inverse-square law (§ 9.46), on the radiation intensity or flux. If operating personnel can be kept at an appreciable distance while the reactor is in operation, a significant saving in thickness and cost of the shielding may be feasible (§ 10.107). For example, a shield may be made thinner at the top and bottom if access is restricted to the sides. Similarly, a smaller thickness may be adequate at the sides and bottom if all operations can be performed from the top.

10.6. The design of a reactor shield usually involves two distinct phases. The initial phase, which is part of the over-all design of the reactor, is concerned with specifying the general configuration of the shield, the materials of construction, and the optimum arrangement that will provide, most economically either in weight or in cost, the required radiation attenuation. It is essential, of course, that the shield be compatible with the general operational requirements of the reactor, and so consideration must be given to the location of the cooling system, control elements, etc. The second phase of shield design is concerned with many details associated with localized problems, such as the effects of ducts and piping that must pass through the shield. As will be seen in the course of this chapter, some of the problems encountered can be solved analytically, but, because of their great complexity, especially those concerned with shielding the reactor itself, recourse must be had to experiment. The conversion of data thus obtained under idealized conditions to actual practical cases is an important aspect of shield design.

Radiations from Reactor Systems

10.7. In principle the radiations which might escape from a reactor system include alpha and beta particles, gamma rays, neutrons of various energies, fission fragments, and even protons resulting from (n, p) reactions. As far as shield design is concerned, however, only gamma rays and neutrons need be considered since these are by far the most penetrating.* Any material which attenuates these radiations to a sufficient extent will automatically reduce all the others to negligible proportions.

* For measurements of gamma-ray spectra from actual reactors, see J. W. Motz, *Phys. Rev.*, 86, 753 (1952); Report AECD-3286; F. Malenschein and T. Love, *Nucleonics*, 12, No. 5, 6 (1954); C. C. Horton, Report AERE-R/R-1271.

10.8. Reactor coolants, spent fuel elements, and solutions of fission products emit relatively few (if any) neutrons, and these are of not very high energy; hence absorption of gamma radiation is, in general, all that need be contemplated. The design of suitable shielding is then a relatively simple matter since, as will be seen shortly, the absence of neutrons eliminates many complicating factors. A special case arises in connection with a circulating fuel system; after leaving the reactor to enter the heat exchanger, the fuel continues to emit delayed neutrons. Although these represent less than 1 per cent of the total fission

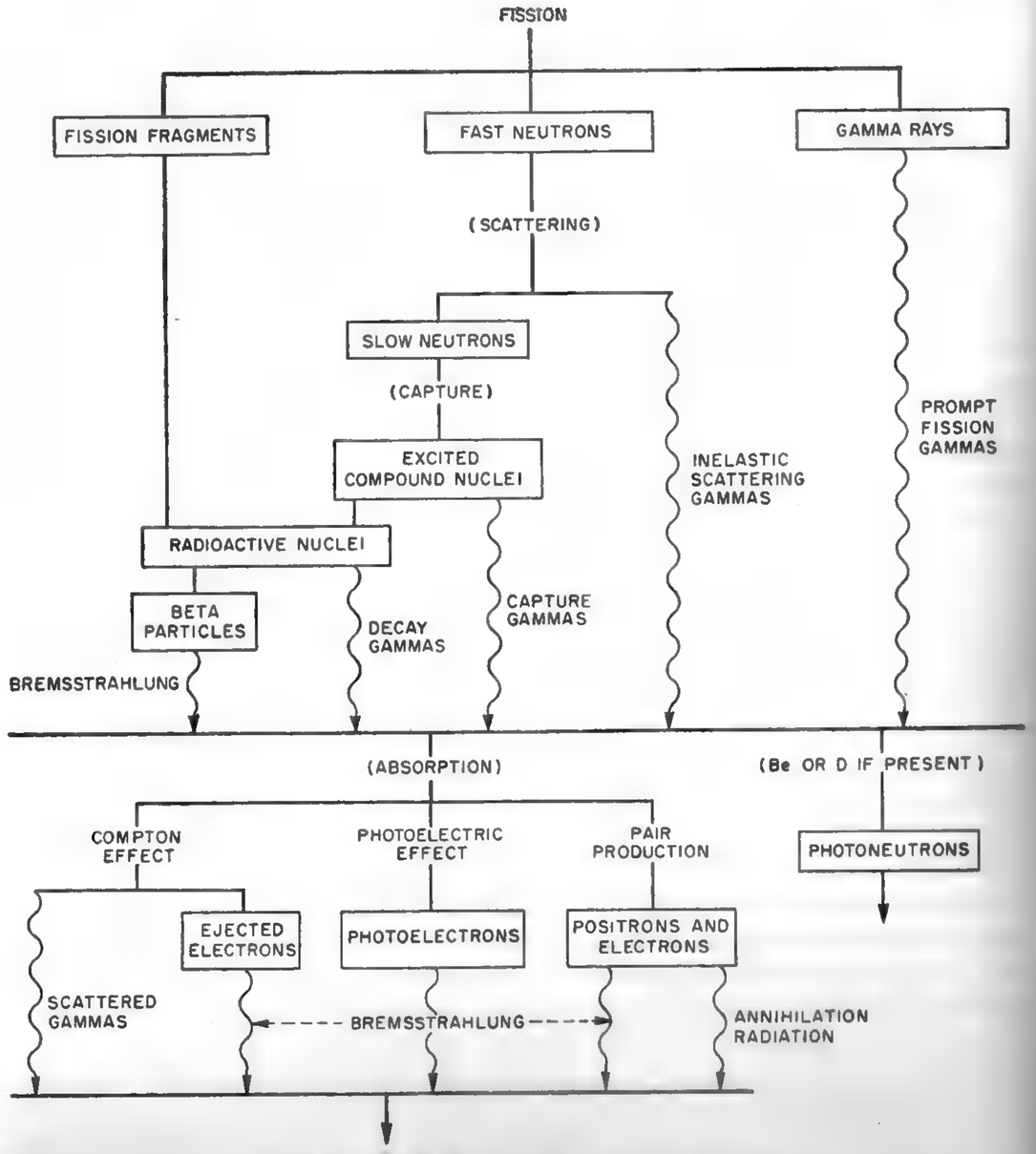


FIG. 10.1. Radiations from nuclear reactor

neutrons and have lower energies than the prompt neutrons, they must definitely not be ignored. In addition to the hazard they themselves represent, they may induce radioactivity, as a result of capture, in the material used as secondary coolant.

10.9. Some indication of the complex character of the nuclear radiations arising from the reactor itself is apparent from Fig. 10.1, which, in accordance with the comment made above, refers only to neutrons and various gamma radiations. The fission process itself produces fission fragments, as indicated at the left of the figure, most of which are radioactive, expelling negative beta particles. Many of these fragments and their decay products emit gamma rays, several of which have energies as high as 2 to 3 Mev. In addition, the slowing down of the beta particles is accompanied by formation of bremsstrahlung (§ 2.24) with a range of energies up to about 2 Mev. The interactions of these various radiations with matter in the reactor produce other radiations which will be considered below.

10.10. More than 99 per cent of the neutrons liberated in fission, indicated in the center of Fig. 10.1, are expelled promptly; as seen in Chapter 2, the energies of these neutrons cover a considerable range (cf. Figs. 2.22 and 2.23). A large proportion of the fission neutrons have energies in excess of 1 Mev, some being as high as 14 Mev, and they are slowed down as a result of elastic collisions with a moderator or by inelastic collisions with the nuclei of heavier elements, e.g., uranium, present in the reactor. As seen in § 2.98, inelastic scattering will be followed by the emission of the excitation energy of the struck nucleus in the form of gamma radiation.

10.11. After being slowed down, many of the neutrons will be involved in (n, γ) reactions. In addition to capture by moderator, coolant, and structural materials, it must be remembered that both uranium-235 and uranium-238 undergo (n, γ) reactions to a very considerable extent. The total energy of the capture gamma rays in each case will be at least equal to the binding energy of the neutron. For the reaction with uranium-238 this energy, as given in Table 2.8, is 5.5 Mev, and for uranium-235 it is about 6.8 Mev. For nuclei of somewhat lower atomic number, the energies of capture gamma rays may be as high as 8 Mev or more.* The products of the (n, γ) reactions are often beta-active, and the slowing down of the beta particles will be accompanied by bremsstrahlung, as noted above. In addition, some of the radioactive species will undoubtedly emit gamma radiation. Finally, the prompt gamma radiation of fission, with a total energy of about 5 Mev divided between two or more photons (§ 2.172), is produced almost simultaneously with the act of fission, as shown at the right in Fig. 10.1.

10.12. If beryllium or deuterium is present in the reactor, either as moderator

* It should be mentioned that, since the total gamma-ray energy is often emitted in two or more quanta of lower energy, the situation is not always as bad as it would at first appear (see Table 2.6).

or reflector, the action of gamma rays of energy exceeding 1.62 or 2.21 Mev, respectively, will produce photoneutrons (§ 2.74).* If sufficient gamma-ray energy is available, some of these photoneutrons will have energies of about 1 Mev or more and will initiate a chain of events similar to that resulting from fission neutrons.

10.13. The interaction of the various gamma radiations will produce photoelectrons, Compton electrons, and, for energies in excess of 1.02 Mev, positron-electron pairs. As these electrons are slowed down, bremsstrahlung will be produced, while the recombination of electrons and positrons will result in the formation of annihilation radiation of energy 0.51 Mev, or possibly 1.02 Mev (§ 2.46).

10.14. The foregoing discussion will suffice to show that the nuclear radiations escaping from a reactor will be very complex in nature; but this is not all. When the neutrons and gamma radiations of various energies enter the shield, they will suffer interactions of the same type as those that occur within the reactor. Throughout the shield there will thus be produced secondary gamma radiations due to inelastic scattering, to (n, γ) reactions, and to other processes involving the nuclei constituting the shielding material. Unless proper precautions are taken in the design of the shield, these secondary components of the radiation could lead to serious hazards. For example, if a slow neutron were captured near the outside of the shield, it could give rise to capture gamma radiation of high energy which might emerge from the shield with little or no energy loss.

10.15. An important matter which must be borne in mind in connection with shield design is the possibility that a radiation component which is negligible within the reactor may become dominant outside the shield. This could happen with fission neutrons of high energy. Because nuclear cross sections, in general, decrease with increasing neutron energy, the removal of fast neutrons is relatively difficult. It is possible, for example, for neutrons of 8-Mev energy or more, which may represent roughly 0.01 per cent of the neutrons within a reactor, to constitute a major proportion of the neutrons leaving the shield. Since the biological effectiveness of neutrons increases with increasing energy, at least up to 3 Mev (cf. Table 9.5), a dangerous situation could arise from these high-energy neutrons alone.

General Aspects of Shield Construction

10.16. In principle the problem of shielding the reactor itself involves three aspects: (1) slowing down the fast neutrons, (2) capturing the slowed-down, or initially slow, neutrons, and (3) absorption of all forms of gamma radiation,

* Actually any nuclear species (except hydrogen) can be made to yield photoneutrons if gamma rays of sufficient energy — equal to the neutron binding energy, i.e., usually more than 6 Mev — are available. Since the cross sections for such (γ, n) reactions are small and photons of 6 Mev (or more) energy are not common in a reactor, photoneutrons emitted by nuclei other than beryllium and deuterium are negligible.

including inelastic-scattering and capture gamma rays, formed by interaction of fast and slow neutrons with nuclei in the shield.

Slowing Down Fast Neutrons

10.17. Since elements of low mass number are the best moderators, it might appear that hydrogen, in the form of water, could best be used as the shield constituent for slowing down neutrons. However, at high neutron energies the scattering cross section of hydrogen is very small, so that considerable thicknesses of water would be required to slow down the fission neutrons of highest energy. The situation can be greatly improved by introducing an element (or elements) of fairly high mass number. Although such substances are not good moderators, i.e., they do not decrease the neutron energy to an appreciable extent in an elastic collision, they are able to reduce the energies of very fast neutrons as a result of inelastic-scattering collisions. Elements such as lead, barium, or iron will readily decrease the neutron energy down to about 0.5 Mev, where the hydrogen (elastic) scattering cross section is several (approximately 4) barns. Hence a combination of a moderately heavy or heavy element with hydrogen will effectively slow down even neutrons of very high energies. The manner in which such combinations can be realized will be considered later.

10.18. It will be seen below that essentially every neutron undergoing an inelastic collision is effectively removed, because of the high probability of subsequent slowing down and capture. Further, even in an elastic collision, when the decrease in energy may not be large, the accompanying change in the direction of motion of the neutron leads to an increased path length through the shield; the chances of slowing down and capture are thereby increased. Consequently it appears, to a first approximation, that the effectiveness of a particular material for the attenuation of fast neutrons is determined by the total fast-neutron cross section, which includes both inelastic and elastic scattering as well as direct capture. As stated in § 2.146, at high energies the total cross section approaches $2\pi R^2$, where R is the nuclear radius. Since R is proportional to $A^{\frac{1}{3}}$, where A is the mass number of the nuclear species (§ 2.147), it follows that the total fast-neutron cross section is proportional to $A^{\frac{2}{3}}$. The number of atoms (or nuclei) in a certain mass of material is inversely proportional to the atomic weight (or mass number). Hence the shielding effectiveness of a given mass of material for fast neutrons may be expected to be inversely related to $A^{\frac{1}{3}}$. In other words, the weight of shielding material required to attenuate fast neutrons to a specified extent should be less for an element of low mass number than for one of high mass number.

10.19. It should be borne in mind, however, that for light elements the threshold energy, below which inelastic scattering of neutrons is not possible, is several million electron volts (§ 2.100). In fact, hydrogen is incapable of causing inelastic scattering at any energy. Light elements would thus not serve the purpose of slowing down fission neutrons by inelastic scattering into the energy

region in which elastic collisions become effective because of the increased cross section. On the other hand, for elements of moderate and high mass numbers the threshold energy for inelastic collisions may be as low as 0.5 Mev or less, so that these elements are able to decrease the energies of very fast neutrons into the region where elastic collisions, e.g., with hydrogen, occur readily. To serve this purpose, therefore, it is desirable that elements of fairly high mass number be included in reactor shields. In comparing two such elements, e.g., iron and lead, the theoretical arguments presented above would indicate that a smaller mass of iron would be as effective as a larger mass of lead for the attenuation of fast neutrons (cf. § 10.114).

10.20. It is worth noting here that the reflector can make an important contribution to fast-neutron shielding. The reflector, especially of a thermal reactor, is invariably a good moderator, e.g., water, heavy water, beryllium, beryllium oxide, or graphite, so that it will slow down an appreciable fraction of the moderately fast neutrons escaping from the reactor core. Because of scattering, many of these slowed down neutrons are returned to the core, and thus cease to be a shielding problem.

Slow-neutron Capture

10.21. The next aspect of reactor shielding to consider is that of capturing the neutrons after they have been slowed down. Actually this is a relatively simple matter, even though the shield does not contain an element with a large or fairly large cross section for the capture of slow neutrons. In fact, it is generally accepted that in a shield almost every neutron which has suffered inelastic scattering may be regarded as removed, since the probability of further slowing down and subsequent capture is large. This is particularly true if a good moderating element, such as hydrogen, is present in addition to one of medium or high mass number.

10.22. The capture cross section (σ_a) of hydrogen for thermal neutrons is only 0.33 barn; nevertheless, the number of these nuclei present in the shield for slowing down purposes is usually sufficient to absorb a large fraction of the neutrons. In addition, iron has an appreciable capture cross section ($\sigma_a = 2.5$ barns) for thermal neutrons. The disadvantage of absorption by iron is that the capture gamma rays have an energy of over 7.5 Mev, essentially all of which is emitted as a single photon; consequently additional gamma-ray shielding becomes necessary. It should be noted in this connection that capture of neutrons by hydrogen is accompanied by 2.2-Mev gamma radiation which can by no means be ignored.

10.23. The difficulty due to capture gamma radiation may perhaps be partly overcome by using lead in place of iron as the inelastic-scattering material. The capture gamma-ray energy, of some 7 Mev, also appears as a single photon, but the capture cross section ($\sigma_a = 0.2$ barn) for lead is less than for iron. It would then be necessary to add to the shield a substance which readily captures slow

neutrons in a reaction other than a (n, γ) process. Such a material is boron-10, which has a large cross section for the (n, α) reaction with slow neutrons (§ 2.138). Although some of the liberated energy does appear as gamma radiation, its energy is only about 0.5 Mev, so that it is readily absorbed. Proposals have been made for incorporating boron in various forms in reactor shields (§ 10.48), especially as its capture cross section is quite appreciable even for fast neutrons.

10.24. Since cadmium captures slow neutrons so readily, and it also has a fairly high mass number, it might be thought that this element could be used as a shielding material. The difficulty here, as with iron, is the energy (about 7.5 Mev divided among two or more photons) of the capture gamma rays. Because of the large neutron capture cross section, the intensity of this radiation would be so high that its absorption would be a serious problem. If, for any reason, a cadmium sheet is used as part of a shield, it should be located near the reactor so that it is backed by adequate material for absorbing the capture gamma rays.

10.25. Attention should be drawn, at this point, to another possible drawback associated with the use in a shield of a material, such as cadmium, having a high resonance peak for the capture of neutrons in a limited energy range (cf. Fig. 2.17). Although most of the neutrons within this range are captured, a large proportion of the others, especially those with higher energies, will penetrate the shield. A hazardous situation might then result from placing too much reliance upon a material that is apparently a very good absorber of neutrons.

Absorption of Gamma Radiation

10.26. The third and final aspect of the shielding problem is the absorption of the various gamma rays, both primary and secondary. As far as their behavior is concerned, the origin of these radiations is not significant; all that matters is their energy and so they are effectively absorbed by a material of high density (§ 2.68). It is evident, therefore, that the shield constituent which serves as the inelastic scatterer of neutrons, i.e., iron or lead, will also function as the absorber of gamma radiation.

10.27. It will be shown below (see also Chapter II) that, within the energy range of interest, the absorption is determined essentially by the mass of the shielding material. This means that the thickness of shield required to produce a specified absorption of gamma rays is inversely proportional to the density of the shielding material. Thus a smaller volume of lead than of iron would be required, but the masses would be approximately the same (see, however, § 10.38).

Summary

10.28. From the foregoing discussion it can be concluded that, in essence, a reactor shield will consist of an element (or elements) of high mass number; one (or more) of low mass number, preferably hydrogen; and possibly, although it

is not mandatory, a good neutron absorber which undergoes (n, α) or (n, p), rather than (n, γ), reactions. The heavy element may be in the metallic state, e.g., iron or lead, or in the form of a compound, e.g., an iron oxide (iron ore) or barium sulfate (barytes). Hydrogen is usually present as water, either as such or in a combined state, although other hydrogen compounds have been considered for use in reactor shields (see § 10.47). If a special neutron absorber is included, it is usually boron in the form of a suitable compound.

10.29. It is appropriate to conclude this section with a note of caution against attempting to design a shield consisting mainly of a material of moderate or high mass number, e.g., lead or iron, even if it has a fairly large cross section for the capture of thermal neutrons. With such a shield neutrons in the intermediate range, with energies too low for inelastic scattering and too high for capture, would not be appreciably attenuated. Even if a material is added outside the shield to slow down and capture these neutrons, the secondary gamma radiation will constitute a hazard unless a considerable thickness is used. A good shield consequently requires the proper combination of moderator (and absorber) with the inelastic scatterer of high mass number.

Shield Configurations

10.30. For maximum efficiency a shield should attenuate fast neutrons and gamma rays at such a rate that the respective fluxes will be reduced to their maximum permissible values (Table 9.6) at the same place, i.e., just at the exterior of the shield. This requirement might be met if the material of high mass number and the hydrogen (or hydrogen compound) were uniformly distributed, in the proper proportions, throughout the shield. Since the heavy material, e.g., iron, is usually a solid, and the hydrogen is generally in the liquid state as water, a completely homogeneous form is not possible. An approximation to uniformity may be achieved by the employment of a vessel of water, possibly containing a dissolved boron compound, in which are a number of parallel sheets of iron (or lead) more or less equidistantly spaced.

10.31. At one time there was much interest in laminated shields for reactors; these consisted of alternate layers of a solid compressed wood product, called Masonite, containing about 6 per cent by weight of hydrogen,* and metallic iron. Later investigations have shown that, although the laminated type of shield is efficient, it is expensive. The same attenuation of neutrons and gamma radiation can be achieved at a much lower cost by incorporating the heavy element, such as iron or barium, in concrete, which normally consists almost entirely of elements of moderate or low mass number, including hydrogen. In this way a fairly uniform distribution of the important constituents is attained, with the added advantage that the concrete can also serve as a structural material. Special "heavy concretes," which will be described in § 10.44, have been extensively used in the shielding of stationary reactors.

* Water contains 11 per cent by weight of hydrogen.

Thermal and Biological Shields

10.32. Essentially all the energy absorbed in the shield from the fast neutrons and gamma rays is ultimately degraded into heat. This means that, for a reactor of moderate or high power, a considerable portion of heat is generated within the shield (see Chapter XI). Since the absorption of both neutrons and gamma radiation is, at least approximately, exponential in character, a large proportion of the total heat liberated will be released in those parts of the shield closest to the reactor. For example, suppose that the gamma-ray flux is 10^{13} photons/(cm^2)(sec) at the inside face of the shield, i.e., nearest the reactor, and this is reduced to 10^3 photons/(cm^2)(sec) at the outer face. If the thickness of the uniform shield is 100 cm, then, assuming an exponential absorption with distance (which will not be strictly the case), it follows that the flux will be decreased from 10^{13} to 10^{12} photons/(cm^2)(sec) within the first 10 cm of the shield. This decrease in flux represents a 90 per cent absorption of the gamma-ray energy in the first 10 per cent of the shield.

10.33. To protect the shield from possible damage from the heat liberated upon absorption of radiation, a so-called *thermal shield* is frequently introduced close to the reactor. It consists of a substantial thickness of a dense metal of fairly high melting point, e.g., iron, placed between the reactor core and the main shield, referred to as the *biological shield*. In the Materials Testing Reactor (MTR), for example, the thermal shield consists of two four-inch thick layers of steel, separated by a space of four inches through which air is passed to serve as coolant.

10.34. It will be noted that the thermal shield consists of a material which is effective for absorbing gamma radiations and for the inelastic scattering of fast neutrons. Since these two types of radiation carry most of the energy leaking from the reactor, it is evident that a large proportion of the heat produced in the shield will be released in the thermal shield. When operating at its full power of 30,000 kw, the MTR produces 48 kw of heat in the thermal shield and about 2 kw in the biological shield made of special concrete.*

10.35. For reactors operating at low or moderate power, the amount of heat liberated in the shield may not be excessive. Further, if the reactor and the shield are large, the local temperatures may not be sufficiently high to cause appreciable harm to the shield. In cases of this kind the introduction of a thermal shield may be unnecessary. The X-10 (natural uranium) reactor at Oak Ridge National Laboratory, for example, does not have a thermal shield, although a special stream of air is directed over the inner faces of the biological shield at the front and back of the reactor. After about ten years of operation, most of the time at an average power level of about 3800 kw, there are no indications of damage to the concrete of the shield.

* J. R. Huffman, *Nucleonics*, 12, No. 4, 21 (1954).

Shield Materials*

10.36. Materials used in shielding may be divided into three broad categories, according to their function: (1) heavy elements to absorb the gamma radiation and slow down very fast neutrons to about 0.5 Mev by inelastic collisions; (2) hydrogenous substances to moderate neutrons having energies in the range below about 0.5 Mev; and (3) materials, notably those containing boron, which capture neutrons without producing high-energy gamma rays. Although these three classes will be considered separately, it will be seen that the same material may often serve two or all three purposes.

Heavy Elements

10.37. The only heavy element which has hitherto been employed in the metallic form for shield construction is iron. In the massive state it has been used for the thermal shield in the MTR, as stated above, and also as a component of the biological shield of the now dismantled Los Alamos Fast Reactor. However, iron turnings or punchings, as well as iron oxides, have been incorporated in concrete for shielding purposes. Further reference to special concretes which incorporate heavy elements, such as iron or barium in some form, will be made shortly.

10.38. Because of its high density and ease of fabrication, lead is being considered as a shield component. For gamma rays with energies in the region of 2 Mev, roughly the same mass of lead as of iron is required to absorb a specified fraction of the radiation. However, at both higher and lower energies the mass-absorption efficiency of lead is appreciably greater than that of iron.

10.39. Disadvantages of lead in reactor shields are its relatively low melting point and its softness. It cannot carry any appreciable portion of the reactor system load and since it cannot withstand high temperatures it would require cooling. The same is true for bismuth, which otherwise might be possible as a shielding material. The elements tantalum and tungsten have both high densities and high melting points, but because of their cost they would be of value only in special cases, for example, as thermal shields in mobile reactors operating at high temperatures. Tungsten has a fairly high radiative capture cross section for thermal neutrons, and the resulting gamma rays must not be overlooked.

10.40. From the long-range point of view, carbides, nitrides, and possibly borides of elements of high mass number may be of interest. Ceramic materials of this type are usually refractory, have high melting points, and are relatively dense, although they may prove difficult to fabricate. The presence of a light element, such as carbon, nitrogen, or boron, would contribute to the slowing down of the fast neutrons, and boron would, in addition, capture neutrons.

* A. E. Pavlish and J. C. Wynd, Report AECD-3007; E. G. Tirpak, Report AECD-3134; *Civil Eng.*, **21**, 451 (1951); T. Rockwell, Report AECD-3352; J. A. Murray and J. H. Cullinan, Report AECU-1835; E. J. Callan, *J. Am. Concrete Inst.*, **25**, 17 (1953).

Hydrogenous Materials

10.41. The value of a hydrogenous material for neutron shielding is determined, first, by its hydrogen content; this is best expressed in terms of the number of hydrogen atoms (or nuclei) per unit volume. In this respect water, with 6.7×10^{22} atoms/cm^3, ranks high, and it has, in addition, the advantage of low cost. However, water as such, because of its liquid nature, has not been greatly used in the past, but it is now attracting interest for research reactors. The low boiling point and instability to radiation must, of course, be taken into consideration.

10.42. Masonite (§ 10.31), with a density of about 1.3 g/cm^3, was used as the hydrogenous material in some of the early reactors. The number of hydrogen atoms per cubic centimeter is as high as about 5.5×10^{22}, which is not much less than that for water. In addition, it contains both carbon and oxygen which can act as moderators. Although it has proved satisfactory in the past, Masonite will probably find little application in the future because of the relatively high cost of erecting laminated shields.

10.43. As a general shield material, there is much to recommend concrete; it is strong, is not expensive, and is adaptable to both block and monolithic types of construction. Ordinary concrete of density 2.3 g/cm^3 contains somewhat less than 10 per cent by weight of water when cured, i.e., about 1.4×10^{22} hydrogen atoms/cm^3. Although this is considerably less than the concentration in water, the larger proportion of oxygen (which acts as additional moderator) and the calcium and silicon in concrete compensate, to a great extent, for the difference. Thus the relaxation length (§ 10.64), which may be regarded broadly as an inverse measure of the shielding effectiveness, for moderately fast neutrons in ordinary concrete is 11 cm, as compared with about 10 cm in water (Table 10.3). Nevertheless, ordinary concrete alone is not very efficient as a reactor shield material since it normally contains no element of high mass number.

10.44. In recent years various special ("heavy") concretes, incorporating heavy elements, have been developed for reactor shielding.* In barytes concrete, for example, the mineral *barytes*, consisting mainly of barium sulfate, largely replaces the sand and gravel aggregate in ordinary concrete. This material was used for the biological shield of the MTR.† The density of barytes concrete is about 3.5 g/cm^3, and the relaxation lengths for fast neutrons and gamma radiation are 8 and 10 cm, respectively.‡ Thus a shield of barytes concrete would have to be no thicker than an iron-water shield of the same effectiveness, although the total weight of the former would be greater.

* It is of interest to record that heavy concretes, incorporating metallic iron or iron ore, were used in construction work for several years before their application to reactor shielding.

† For description of construction of MTR shield, see L. Narrow, *Eng. News-Record*, **152**, No. 10, 36 (1954).

‡ The relaxation lengths of lead and iron for gamma rays of 3 to 5 Mev energy are about 2.5 and 3.7 cm, respectively; that of water is 30 cm.

10.45. The barytes aggregate may be partly or wholly replaced by *limonite*, an iron ore of composition $2Fe_2O_3 \cdot 3H_2O$; this contributes both iron and bound water to the concrete. If desired, iron punchings may be added to the resulting concrete. Thus the Brookhaven reactor shield is made of a mixture of iron ore, iron punchings, Portland cement, and water; it has a density of 4.27 g/cm³, and the relaxation lengths for fast neutrons and gamma rays are 6.3 and 8 cm, respectively. The compositions (by weight) of two fairly typical concretes for use in reactor shielding are given in Table 10.1.

TABLE 10.1. COMPOSITION BY WEIGHT OF CONCRETES FOR REACTOR SHIELDS

Barytes Concrete (*Density* 3.5 g/cm³)		*Iron Concrete* (*Density* 4.5 g/cm³)	
Barytes	60%	Steel punchings	57%
Limonite	22%	Limonite	26%
Portland cement	11%	Portland cement	13%
Water	7%	Water	4%

10.46. Various other heavy cements and concretes have been devised; one, consisting of iron, Portland cement, and water, has a density as high as 6 g/cm³. Unless it is kept soaked with water, which is a definite possibility, the hydrogen content of this material might be too low for it to serve as a neutron shield. The total amount of water held by concrete when cured can be increased by adding certain water-absorbing substances, but these usually have a fairly low density. Boron compounds, e.g., the mineral *colemanite* ($2CaO \cdot 3B_2O_3 \cdot H_2O$), have also been incorporated into concretes in order to increase the probability of neutron capture without high-energy gamma-ray production.

10.47. As possible alternatives to water as the hydrogenous component of a reactor shield, some consideration has been given to metal hydrides, since they possess features of special interest, especially for high-temperature shields. Such materials would combine both inelastic and elastic neutron scattering with gamma-ray absorption properties. Two hydrides, in particular, have been studied: titanium hydride (TiH_2) and zirconium hydride (ZrH_2), which contain approximately 9.0×10^{22} and 7.5×10^{22} hydrogen atoms/cm³, respectively. At high temperatures, e.g., above 500°C (930°F), these compounds decompose, giving off hydrogen, but it appears that the dissociation can be greatly diminished by the presence of a small excess of the metal. Other heavy-metal hydrides, about which little is yet known but which have been mentioned as worthy of consideration, are those of chromium, iron and related metals, tungsten, thorium, and uranium.

Boron Compounds

10.48. There are several common boron compounds which are sufficiently soluble to be added to a water shield to increase the rate of neutron capture; among these, mention may be made of boric acid, borax, and various other borates. The advantage to be gained by the boration of water, however, varies considerably with the character of the shield. In a lead-water shield, with a large proportion of the lead near the core, for example, it would be beneficial to add boron to the water. The fact that there is relatively little gamma-ray absorbing material in the outer regions of the shield would then not be serious since only low-energy gamma rays are produced when boron captures neutrons.

10.49. A boron-containing material which may prove of value in reducing the dimensions, and hence the over-all cost, of concrete shields is *boral*, a complex of boron carbide (B_4C) and aluminum.* The two powdered constituents, in the proportions of roughly 45 to 55 by weight, are mixed and heated. The product is then rolled down to a thickness of about 0.25 in., and the resulting sheet is clad on each side with about 0.02 in. of aluminum. The product has a density of 2.5 g/cm³; it has satisfactory mechanical properties and absorbs neutrons with no accompanying high-energy gamma radiation. A similar material which has been proposed, called *boraxal*, is made from aluminum and boric oxide (B_2O_3). It is much cheaper than boral but contains somewhat fewer boron atoms per unit volume.

SHIELDING GEOMETRY

The Point Kernel

10.50. For many aspects of shield design it is useful to have equations which give the intensity of the radiation, received at a detector, from sources of various shapes, e.g., point, linear, planar, spherical, etc., as a function of the distance between source and detector, i.e., of the shield thickness. Such equations can be applied directly to calculate the thickness of shield required to reduce the radiation intensity at the detector to a specified amount. Alternatively they can be used to determine the intensity distribution from a source of one particular shape and size by employing experimental measurements made with a different source. Both types of application of what is frequently called *shielding geometry* will be described in this chapter. The results obtained are quite general and are equally applicable to neutrons and to gamma radiation.

10.51. The calculation of the radiation intensity due to a distributed source, i.e., a linear, surface, or volume source, as a function of distance makes use of the fundamental principle that any such source can be treated as a distribution of point sources. The first step in the derivation of the required equations is to write an expression for the radiation intensity received by a point detector at a certain distance from a point source of known strength. The resulting expression is then integrated over the whole of the distributed source. In some cases the required integration can be carried out analytically, but in others either approximations must be made or special methods of integration used.

* V. L. McKinney, *et al.*, Report AECD-3476.

10.52. For the present purpose it is convenient to define a *point kernel*, $G(R)$, which represents the response of a detector located at a distance R from a unit point source, i.e., a source emitting one particle of radiation (one neutron or one photon) per second, both source and detector being located in an infinite homogenous medium. For a point source of strength S, the intensity (or flux) of the radiation received at a detector at a distance R from the source is given by $SG(R)$, i.e., S times the value for a unit point source.

10.53. The actual form of $G(R)$ will depend on the nature of the medium, i.e., the shield, separating the detector from the source as well as on the character, e.g., type and energy, of the radiation. For simplicity it will be assumed that $G(R)$ is a function only of the distance R between the source and the detector, i.e., of the shield thickness, for a given radiation and shielding medium. In other words, for any specified value of R, the medium is supposed to produce the same attenuation of the radiation, irrespective of position or direction.* This implies, among other things, an isotropic source, i.e., one which radiates uniformly in all directions, and also an isotropic shielding medium. A laminated shield (§ 10.31), for example, would not satisfy the latter condition, but the use of the function $G(R)$ is, nevertheless, a convenient approximation.

Distributed Radiation Sources

10.54. In the theoretical treatment of extended (or distributed) radiation sources it is necessary, if the results are to be unambiguous, to specify the directional character of the detector. Two hypothetical idealized types are used in calculations: directional and isotropic. Most actual radiation detectors, including the human body, resemble both types to some extent, although the similarity is usually greater for one than for the other.

10.55. The *directional detector* is assumed to measure only the component of the radiation in a particular direction, so that it indicates (per cm² per sec) the radiation current (§ 3.12). The *isotropic detector*, on the other hand, collects radiation from all directions, and so provides a measure of the radiation flux. Such a detector will always record an intensity of radiation at least as high as that indicated by the directional detector and may be regarded as an upper limit to the radiation that would be received by a human body. The reading of the hypothetical isotropic detector would thus provide a basis for conservative shield design, and in the calculations given below the use of this type of detector will be postulated. Since the response of the isotropic detector, in terms of neutrons or photons received per second, can be used to derive an approximate measure, at least, of the radiation dose rate, it will be represented by the symbol D.

* It will be seen below that an expression for the function $G(R)$ which is frequently used is similar to equation (9.49.1) for gamma radiation. It is based on exponential absorption and inverse-square-law attenuation with distance.

Isotropic Circular Plane Source

10.56. The first case of a distributed source to be considered is that of an isotropic circular plane (disk) source of radius a, with the detector on the normal passing through the center (Fig. 10.2). The source strength S is the total number of particles emitted (isotropically) per unit area per second. Consider a narrow annulus of radius ρ and width $d\rho$; the area of this annulus is $2\pi\rho\, d\rho$ and it is everywhere distant R from the detector. The radiation received per second at the detector is equivalent to that from $2\pi\rho\, d\rho$ point sources of strength S, namely, $2\pi\rho\, d\rho\, SG(R)$, where $G(R)$ is the point kernel defined above.

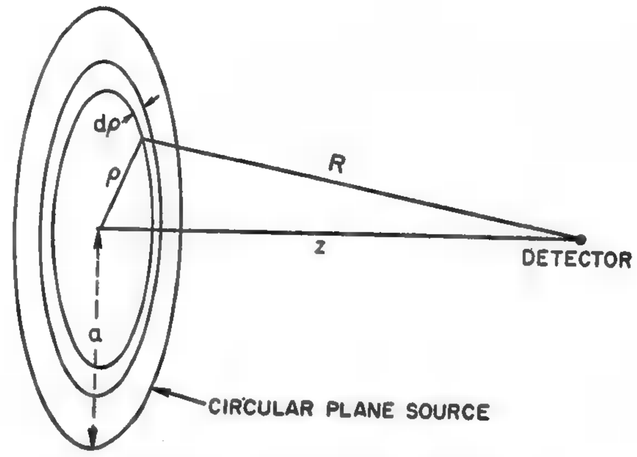


Fig. 10.2. Dose from circular plane source

10.57. The corresponding value for the whole disk source is then obtained by integrating over all values of ρ from zero to a; the result is

$$D = 2\pi S \int_0^a G(R)\rho\, d\rho.$$

Since $R^2 = \rho^2 + z^2$, where z is the thickness of the shield, it follows that

$$2R\, dR = 2\rho\, d\rho$$

and hence

$$D = 2\pi S \int_z^{\sqrt{z^2+a^2}} G(R)R\, dR, \qquad (10.57.1)$$

the limits of integration having been changed to correspond to the new variable. For a large disk the upper limit may be set equal to infinity, thus simplifying the integration; then

$$D = 2\pi S \int_z^{\infty} G(R)R\, dR, \qquad (10.57.2)$$

which is strictly applicable to an infinite plane source.

Isotropic Spherical Surface Source

10.58. A case of interest, which has a number of important applications, is that of an isotropic source that is spread uniformly over a spherical surface. In order to simplify the treatment, it is postulated that the isotropic absorbing medium extends throughout the inside, as well as the outside, of the sphere. Radiation coming from the surface of the sphere farthest from the detector will thus pass through a single medium, and $G(R)$ will be the same function for all values of R.

10.59. Let r be the radius of the spherical surface and r_0 the distance from the center of the sphere to the detector (Fig. 10.3). Consider a thin annulus, as shown in the figure, all points of which are at a distance R from the detector. The circumference of this annulus is $2\pi r \sin\theta$, and the width is $r\,d\theta$, so that its

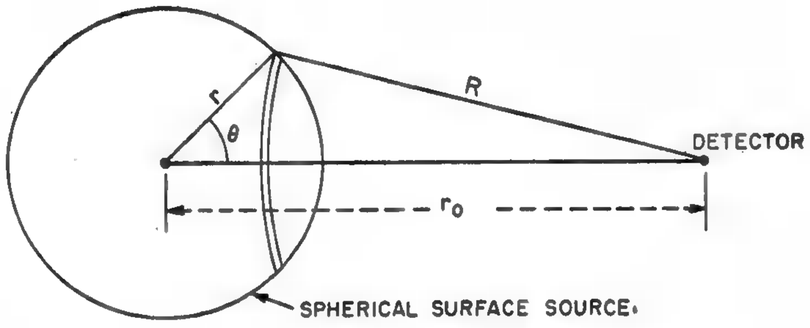


FIG. 10.3. Dose from spherical surface source

area is $2\pi r^2 \sin\theta\,d\theta$. If S is the source strength, expressed as particles emitted per unit area per second, it is seen that for an isotropic detector the radiation received per second from the whole spherical surface is

$$D = 2\pi r^2 S \int_0^\pi G(R) \sin\theta\,d\theta. \tag{10.59.1}$$

The relationship between R, r, and r_0 is given by

$$R^2 = r^2 + r_0^2 - 2rr_0\cos\theta,$$

so that

$$2R\,dR = 2rr_0 \sin\theta\,d\theta.$$

Hence, upon changing the variables from θ to R, equation (10.59.1) becomes

$$D = 2\pi S \frac{r}{r_0} \int_{r_0-r}^{r_0+r} G(R) R\,dR. \tag{10.59.2}$$

If $r_0 + r$ is large so that, as an approximation, the upper limit of the integration may be taken as infinity, the integral in equation (10.59.2) will have the same form as that in equation (10.57.2), and analytical solution is generally possible.

10.60. In any event, the integral in equation (10.59.2) may be expressed as the difference between two integrals: one with the limits from $r_0 - r$ to infinity and the other from $r_0 + r$ to infinity. Consequently,

$$D = \frac{r}{r_0}\left[2\pi S \int_{r_0-r}^{\infty} G(R) R\,dR - 2\pi S \int_{r_0+r}^{\infty} G(R) R\,dR\right]. \tag{10.60.1}$$

Upon comparison with equation (10.57.2), it is seen that each integral is equivalent to the dose rate from an infinite plane source, the first as observed at a distance $r_0 - r$ and the second at $r_0 + r$. Hence, for the same source strengths per unit area, equation (10.60.1) can be written as

$$D(\text{sphere}) = \frac{r}{r_0}\,[D(\text{inf. plane at } r_0 - r) - D(\text{inf. plane at } r_0 + r)], \tag{10.60.2}$$

so that the dose rate from the spherical source can be expressed in terms of the values for two infinite planar sources. If the diameter of the sphere is relatively large, the second term on the right of equation (10.60.2) is negligible, and this equation becomes

$$D(\text{sphere}) \approx \frac{r}{r_0} D(\text{inf. plane at } r_0 - r). \tag{10.60.3}$$

Thus the dose rates to be expected from a spherical source can be calculated from the data for an infinite plane source, or vice versa. It is of interest to note that equation (10.60.3) implies that the radiation intensity received at the detector from the spherical source, after passage through a given shield, is less by a factor of r/r_0 than that due to an infinite plane source (of the same strength per unit area) located at the distance of closest approach, i.e., $r_0 - r$, from the detector.

Isotropic Cylindrical Surface Source

10.61. The derivation will be given later (§ 10.144), but it is of interest, for the sake of completeness, to quote here the expression giving the radiation received from an isotropic cylindrical surface source of infinite length. If r is the radius of the cylinder and r_0 the distance of its axis to the detector (Fig. 10.4), then

$$D(\text{cylinder}) \approx \sqrt{\frac{r}{r_0}}\, D(\text{inf. plane at } r_0 - r). \tag{10.61.1}$$

Since r/r_0 is here less than unity, $\sqrt{r/r_0}$ is greater than r/r_0; the radiation dose rate from an infinite cylindrical source thus lies between that due to an infinite plane source, on the one hand, and that due to a large spherical source having the same radius as the cylinder, on the other hand. The source strengths per unit area are assumed to be the same in all cases.

10.62. Although equation (10.61.1) applies to an infinite cylinder, it is a good approximation for any cylinder that is not very short in comparison with the

distance from the detector to the nearest point on the cylinder. The physical reason for this is that points on the cylinder that are distant from the detector contribute relatively little to the radiation dose. Consequently the radiation received from an infinite cylinder would be very little more than from a long finite cylinder. However, if the cylinder is short compared with the distance to the detector, special methods must be used.

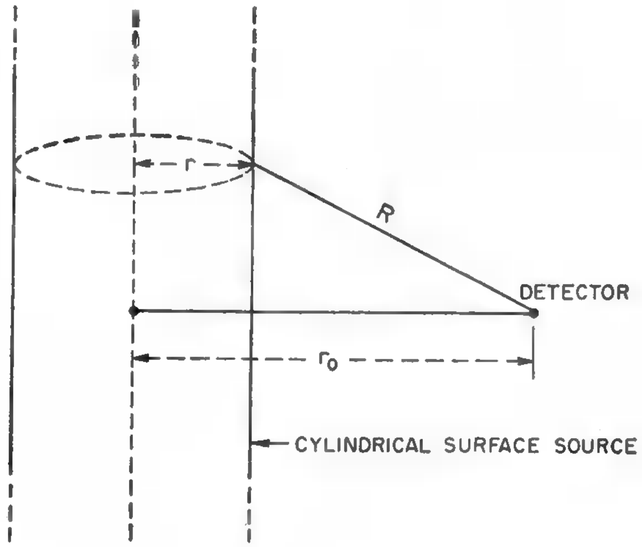


FIG. 10.4. Dose from cylindrical surface source

Example After traversing a shield 6 ft thick, the radiation dose received from a large cubical reactor was found to be equivalent to 1 mrem/hr. Estimate the corresponding dose rates for the same shield if the reactor were (1) a sphere 3 ft in radius and (2) a long cylinder with a radius of 3 ft. The surface source strength, per unit area, is assumed to be the same in all cases.

(1) For the spherical surface source, r is 3 ft and r_0 is $3 + 6 = 9$ ft (see Fig. 10.3). Since the sphere is relatively large, equation (10.60.3) is a good approximation, so that

$$D(\text{sphere}) \approx \tfrac{1}{8}D(\text{inf. plane at 6 ft}).$$

The face of the large cubical reactor behaves essentially as an infinite plane and since the dose rate received from it after passage through the 6-ft shield is 1 mrem/hr, the dose rate to be expected from the spherical reactor (having the same surface source strength per unit area) is 0.33 mrem/hr.

(2) For the long cylinder, r is 3 ft and r_0 is 9 ft (see Fig. 10.4); hence, from equation (10.61.1),

$$D(\text{cylinder}) \approx \sqrt{\tfrac{1}{8}}\, D(\text{inf. plane at 6 ft})$$
$$= 0.57 \text{ mrem/hr}.$$

ABSORPTION AND GEOMETRIC ATTENUATION OF RADIATION

10.63. The expressions derived above are completely general in character. They are independent of the nature of the radiation and also of the nature of

the point kernel $G(R)$ which gives the dose rate received from a point source as a function of R, its distance from the detector. However, in order to make use of the results for shielding calculations it is necessary to specify $G(R)$ explicitly. As seen in Chapter II, absorption of both neutrons and gamma rays of a given energy can be represented by an exponential expression of the form

$$I(R) = I(0)e^{-\Sigma R}, \tag{10.63.1}$$

where $I(0)$ and $I(R)$ are, respectively, the radiation intensities before and after passage through a thickness R of shield, and Σ is the macroscopic absorption cross section for the radiation. For gamma rays Σ is equivalent to μ, the linear absorption coefficient, as pointed out in § 2.116.

10.64. In reactor shielding the radiations to be absorbed are not monoenergetic and, in any event, because of energy degradation in scattering processes, the energy distribution of both neutrons and gamma-ray photons does not remain constant throughout the thickness of the shield. Hence, if an exponential absorption equation, of the form of (10.63.1), is to be used, the cross sections will generally have no precise significance apart from their empirical application to a particular set of circumstances. For calculational purposes it is convenient to retain the exponential character of the absorption equation, but to modify it by writing

$$I(R) = I(0)e^{-R/\lambda}, \tag{10.64.1}$$

where $I(R)$ and $I(0)$ have the same significance as before and λ, which is equivalent to the mean free path (§ 2.122), i.e., to $1/\Sigma$ (or $1/\mu$), for monoenergetic neutrons or photons, is called the *relaxation length* of the shield material for the given radiation. It can be seen from equation (10.64.1) that, physically, it is the distance (or thickness) of shield in which some property of the radiation, e.g., the intensity or flux, is decreased by a factor of $1/e$, i.e., 1/2.72.

10.65. The relaxation length is frequently used in shielding work whether the radiation absorption is exponential or not; it may be defined, quite generally, by

$$\frac{1}{\lambda} \equiv -\frac{d \ln I(R)}{dR} = -\frac{1}{I(R)} \cdot \frac{dI(R)}{dR} \tag{10.65.1}$$

at a point, described by R, in a given medium. Then, if $I(R)$ is an exponential function of R, as in equation (10.64.1), the value of λ is constant over the exponential range. But, if the absorption is not exponential, the relaxation length will vary from point to point in the medium.

10.66. The simple exponential absorption equation (10.64.1) is strictly applicable only to narrow collimated (parallel) beams of radiation since it is based on the postulate that scattered particles are completely removed from the radiation beam. In other words, the scattered radiation can be treated effectively as if it were absorbed. However, for a relatively thin layer of absorber, i.e., for a thin shield, the exponential equation does represent a good approximation,

even if the beam is not well collimated. This is because the increased path length and the change in direction of motion after scattering greatly reduce the probability that the radiation will reach the detector (Fig. 10.5). On the other hand, if the shield is relatively thick, some particles which have suffered two or more scattering collisions within the absorber may reach the detector (Fig. 10.6). In this case the scattered particles are not removed, and the intensity at the detector as calculated from the exponential equation will be too low.

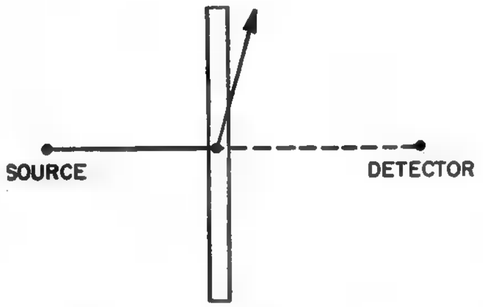


FIG. 10.5. Scattering of radiation in thin shield

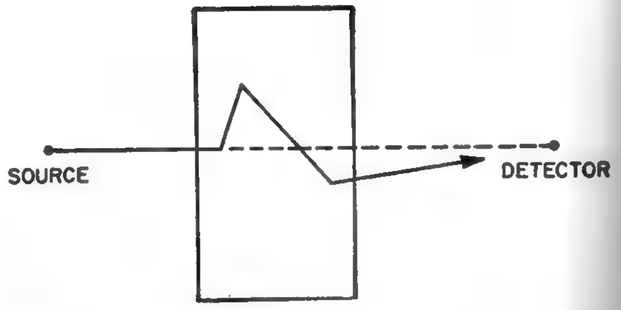


FIG. 10.6. Scattering of radiation in thick shield

10.67. The effect of scattered radiation is allowed for by means of what is called a *build-up factor*, $B(R)$, which is usually taken to be a function of the shield thickness. The exponential absorption equation then takes the general form

$$I(R) = B(R)I(0)e^{-R/\lambda}, \tag{10.67.1}$$

where $B(R)$ depends on the energy of the neutrons or gamma-ray photons. The value of $B(R)$ is always greater than unity, but for thin shields, especially when R/λ (or μR) < 1, the build-up factor is approximately unity, i.e., $B(R) \approx 1$. For gamma-ray absorption in thick shields, when R/λ (or μR) $\gg 1$, the build-up factor is very roughly equal to R/λ (or μR), i.e., to the number of relaxation lengths in the shield thickness; this is referred to as a *linear build-up factor*. For fast neutrons, in a water shield at least, the build-up factor is not as large as this, because there is a considerable probability that a scattered neutron will not travel far before it is captured. The foregoing conclusions may be summarized as follows:

When R/λ (or μR) < 1, $B(R) \approx 1$, for fast neutrons and gamma rays
When R/λ (or μR) $\gg 1$, $B(R) \approx R/\lambda$ (or μR) for gamma rays
$B(R) < R/\lambda$ for fast neutrons.

10.68. Regardless of whether a shield is thick or thin, in calculating the radiation intensity at a detector, allowance must be made for the geometric attenuation resulting from the separation of the detector from the source. If a point source emits S particles (photons or neutrons) per sec, then, assuming isotropic distribution, the flux at a distance R cm, disregarding absorption, will be $S/4\pi R^2$ particles/(cm^2)(sec), since $4\pi R^2$ is the area of a spherical surface at R (cf. § 9.45). By combining this result with equation (10.67.1) to allow for absorption of radiation by the shielding medium separating the detector from the source, it follows that the radiation flux, D, at the detector will be given by

$$D = SB(R)\frac{e^{-R/\lambda}}{4\pi R^2}, \tag{10.68.1}$$

where $I(0)$ has been replaced by the source strength, S, to which it is equivalent.

10.69. The right side of equation (10.68.1) gives the flux received at a detector distant R from a point source. It is consequently equal to $SG(R)$, as derived in (§ 10.52), and the expression for the point kernel, $G(R)$, based on exponential absorption combined with a build-up factor, is thus

$$G(R) = B(R)\frac{e^{-R/\lambda}}{4\pi R^2}. \tag{10.69.1}$$

This form of $G(R)$ will be used in some subsequent calculations.

RADIATION FROM INFINITE PLANE SOURCE

10.70. The use of some of the equations obtained above will be illustrated by considering an isotropic plane source.* If this is large, equation (10.57.2) is applicable with the point kernel, $G(R)$, expressed by equation (10.69.1). The two general cases, which lend themselves to analytical treatment, namely, with the build-up factor, $B(R)$, either (1) unity, or (2) linear, will be examined.

Build-up Factor Unity

10.71. In this case $B(R)$ is unity and combination of equations (10.57.2) and (10.69.1) gives for an infinite plane source

$$D = 2\pi S_a \int_z^\infty \frac{e^{-R/\lambda}}{4\pi R^2} R\, dR$$
$$= \frac{S_a}{2}\int_z^\infty \frac{e^{-R/\lambda}}{R}\, dR,$$

where S_a is the surface source strength, expressed in any convenient units per cm^2 per sec. A new variable, x, defined by $x \equiv R/\lambda$, is now introduced, so that

$$D = \frac{S_a}{2}\int_{z/\lambda}^\infty \frac{e^{-x}}{x}\, dx. \tag{10.71.1}$$

The integral in equation (10.71.1) is a form of what is known as the *exponential integral function*, defined by

* For nomograms which can be used for calculations involving sources of various shapes, see J. L. Balderston, J. J. Taylor, and G. J. Brucker, Report AECD-2934.

$$E_n(q) \equiv \int_q^\infty \frac{e^{-p}}{p^n}\,dp, \tag{10.71.2}$$

values of which may be found in tables.* In the present case n is unity and q is equal to z/λ, so that

$$D = \tfrac{1}{2}S_a E_1\left(\frac{z}{\lambda}\right), \tag{10.71.3}$$

where $E_1(z/\lambda)$ is the value of the first order exponential integral function for the argument z/λ. Hence, for an isotropic infinite plane source of strength S_a, the rate of penetration of radiation through a shield of thickness z can be obtained if λ, the relaxation length of the radiation in the shield material, is known, by ascertaining the appropriate value of $E_1(z/\lambda)$ from tables. If direct experimental values of the relaxation length are not available for the medium, i.e., the shield, separating the radiation source from the detector, λ may be taken as equal to $1/\mu$ for gamma rays or to $1/\Sigma$ for neutrons.

Example A large isotropic plane source of fast neutrons emits these particles at the rate of S neutrons/(cm²)(sec). Estimate the dose rate behind a shield of ordinary concrete 7 ft thick, assuming a build-up factor of unity.

Since the plane source is large it may be treated as infinite. The relaxation length of fast neutrons in ordinary concrete is approximately 11 cm (see Table 10.3), and since 7 ft is about 213 cm, i.e., $z = 213$ cm, it follows from equation (10.71.3) that the rate of escape of neutrons from the shield is given by

$$D = \frac{S}{2}E_1\left(\frac{213}{11}\right) = \frac{S}{2}E_1(19.3) \text{ fast neutrons/(cm}^2\text{)(sec)}.$$

From tables of exponential integral functions it is found that $E_1(19.3)$ is close to 2.05×10^{-10} and so D is $1.03 \times 10^{-10}S$ neutrons/(cm²)(sec). (It should be noted that the 7-ft thick shield of concrete attenuates fast neutrons by a factor of about 10^{-10}.)

10.72. When q, in equation (10.71.2), is large, the exponential integral function reduces to a simple form, namely,

$$E_n(q) \approx \frac{e^{-q}}{q^n},$$

so that

$$E_1(q) \approx \frac{e^{-q}}{q}.$$

Consequently when z/λ is large, i.e., for relatively thick shields, equation (10.71.3) reduces to

$$D = \frac{S_a\lambda}{2z}\,e^{-z/\lambda}. \tag{10.72.1}$$

Example Recalculate the data of the previous example using the simple approximation of taking z/λ to be large.

* See, for example, "Tables of Sines, Cosines, and Exponential Integrals," Federal Work Agency Project, WPA, U. S. Government Printing Office, 1940.

Since z/λ is 19.3, it follows from equation (10.72.1) that

$$D = \frac{S}{(2)(19.3)}\,e^{-19.3}$$
$$= 1.0 \times 10^{-10}S \text{ neutrons/(cm}^2\text{)(sec)}.$$

The difference between this result and that obtained using the actual value of the exponential integral function is negligible for the present purpose.

10.73. In most practical situations, z/λ is large; that is to say, the shield is several, e.g., 20 or more, relaxation lengths in thickness. In these circumstances the limiting equation (10.72.1) is completely adequate for many shielding calculations.

Linear Build-up Factor

10.74. When the build-up factor is linear the point kernel is given by equation (10.69.1) with $B(R)$ equal to R/λ, so that for an infinite plane source equation (10.57.2) becomes

$$D = 2\pi S_a \int_z^\infty \frac{R}{\lambda}\cdot\frac{e^{-R/\lambda}}{4\pi R^2}\,R\,dR$$
$$= \frac{S_a}{2\lambda}\int_z^\infty e^{-R/\lambda}\,dR$$
$$= \frac{S_a}{2}\,e^{-z/\lambda}. \tag{10.74.1}$$

10.75. If this expression is compared with equation (10.72.1), for the case of a thick shield with a build-up factor of unity, it is seen that a linear build-up factor results in an increase in the radiation intensity by a factor of z/λ. As seen in the examples given above, this may well be about 20 or so.

10.76. For thick gamma-ray shields the linear build-up factor usually represents a good approximation, and equation (10.74.1) may be used for purposes of calculation. For neutrons, however, the build-up factor is probably less than z/λ, as seen earlier. The actual value of the dose rate, D, will then be somewhere between the values given by equations (10.72.1) and (10.74.1). However, since it is desirable to be conservative in shielding calculations, i.e., to overestimate rather than to underestimate the shielding requirements, it is probably best to use equation (10.74.1) for neutrons, as well as for gamma rays.

Finite Plane Isotropic Source

10.77. In the foregoing treatment the plane source has been assumed to be large, so that it may be treated as being infinitely large. If this approximation is not justifiable, equation (10.57.1) for a circular plane source of finite size should be used. This may be expressed as the difference between two integrals, the limits of integration being z and infinity for one, and $\sqrt{z^2 + a^2}$ and infinity for the other. Analytical solutions are then possible for the two cases considered

above, in which the build-up factor is either unity or linear. Although the results so obtained apply to a circular plane source, they represent a fair approximation for a rectangular source of the same area.

10.78. A simple, but somewhat approximate, procedure may be employed to estimate the dose rate to be expected from a finite plane source if that from an infinite source is known.* If $D(\infty)$ is the dose rate calculated for an infinite plane source, then $D(a)$ for a plane circular source of radius a, having the same source strength per unit area, is related to it by

$$1 + \alpha > \frac{D(\infty)}{D(a)} \geq \tfrac{1}{2} + \alpha \tag{10.78.1}$$

where

$$\alpha \equiv \frac{2\lambda^2}{a^2}\left(\frac{z}{\lambda} + 1\right). \tag{10.78.2}$$

In general, the lower limit, i.e., $\frac{1}{2} + \alpha$, is a good approximation to the ratio of the radiation dose rates received from infinite and finite sources after penetration through a shield z in thickness.

Example Compare the fast-neutron dose rate to be expected from an infinitely large plane source with that from a circular source of radius 2 ft, of the same strength per unit area, after penetration through a concrete shield 7 ft thick.

The value of a is 2 ft, i.e., 61 cm; z is 213 cm, as before, and for fast neutrons in concrete λ is 11 cm. Consequently, by equation (10.78.2),

$$\alpha = \frac{(2)(11)^2}{(61)^2}\left(\frac{213}{11} + 1\right) = 1.3.$$

The required ratio, which may be taken as roughly equal to $\frac{1}{2} + \alpha$, is thus about 1.8.

10.79. A factor of two is generally within the limits of the errors involved in approximate shielding calculations of radiation dose rates. So for many purposes the treatment of a finite disk with a radius exceeding 2 ft as an infinite plane source does not seriously overestimate the amount of radiation which reaches a detector after passage through a specified shield. Physically, the reason for this is that most of the radiation reaching the detector from a plane source originates from the central region nearest to the detector. The more distant parts of the plane contribute relatively little and their inclusion in calculations, for simplicity, does not usually introduce a significant error.

Spherical Surface Source

10.80. The calculation of the radiation received from a spherical surface source after passage through a shield may be performed by combining the general equation (10.59.2) with the specific expression for the point kernel. In general, however, it is satisfactory to use the relationship, expressed by equation (10.60.2),

* A more exact treatment, of which the result given here is an approximation, will be found in the appendix to this chapter.

between the radiation dose rates to be expected from spherical and (infinite) plane surface sources of the same strength per unit area.

Example Fast neutrons escape from a spherical surface source of 1 ft diameter at the equivalent isotropic rate of S neutrons/(cm²)(sec). This surface, which may be that of a spherical reactor, is surrounded by a graphite reflector 2 ft thick. Assuming a linear build-up factor, estimate the rate at which fast neutrons escape from the reflector.

Referring to Fig. 10.3, it is seen that in this case r is 0.5 ft and r_0 is 2.5 ft; hence $r_0 - r$ is 2 ft, i.e., 61 cm, and $r_0 + r$ is 3 ft, i.e., 91.5 cm. According to equation (10.60.2),

$$D(\text{sphere}) = \frac{0.5}{2.5}[D(\text{inf. plane at 61 cm}) - D(\text{inf. plane at 91.5 cm})].$$

Since a linear build-up factor has been postulated, the infinite-plane dose rates are given by equation (10.74.1). The relaxation length for fast neutrons in graphite may be taken as equal to the reciprocal of the total macroscopic absorption cross section which is about 0.11 cm^{-1}; hence λ is roughly 9 cm. It follows, therefore, that

$$D(\text{sphere}) = \frac{1}{5} \cdot \frac{S}{2}(e^{-61/9} - e^{-91.5/9})$$
$$= 10^{-4}S \text{ neutrons/(cm}^2\text{)(sec)}.$$

(It is evident that the second exponential is negligible in comparison with the first, and so it would have been sufficient to use equation (10.60.3) in this instance. The same would be true for all cases in which the diameter of the sphere is appreciably greater than the relaxation length of the radiation in the shield.)

Volume-distributed Source with Self-absorption

10.81. The discussion so far has referred to surface-distributed sources, although in practice most radiation sources are distributed throughout a volume, e.g., a reactor or a vessel containing radioactive material. However, it will now be shown that the radiation to be expected from a volume-distributed source can be expressed in terms of the flux at the surface. Since there is some self-absorption of the radiation within the volume itself, appropriate allowance must be made, as will be shown below.

10.82. Imagine a large volume-distributed source having a uniform strength of S_v particles (neutrons or photons)/(cm³)(sec), and consider the annular volume element, shown in Fig. 10.7, at a distance R from the point P on the surface of the source and lying on the axis of the annulus. The radius of the annular element is $R \sin\theta$, and its cross sectional area is $(R\,d\theta)(dR)$, so that the volume is $(2\pi R \sin\theta)(R\,d\theta)(dR)$, i.e., $2\pi R^2 \sin\theta\,d\theta\,dR$. The total rate of emission of radiation from this volume element, in particles per sec, is then obtained upon multiplying the volume by S_v. Since all regions of the annulus are distant R from the point P, the flux at the surface due to radiation originating in the annulus will be

$$\text{Radiation flux at surface arising from annular volume} = 2\pi S_v G(R) R^2 \sin\theta\,d\theta\,dR,$$

where $G(R)$ is, as before, the point kernel. The total radiation flux at the surface due to the whole (infinitely large) volume source is now given by integrating this expression over all values of R, from zero to infinity, and all values of θ, from zero to $\pi/2$; thus,

$$\text{Total radiation flux at surface} = 2\pi S_v \int_0^{\infty} \int_0^{\pi/2} G(R) R^2 \sin\theta \, d\theta \, dR. \quad (10.82.1)$$

This quantity has the same value at all points on the surface of the large volume-distributed source.

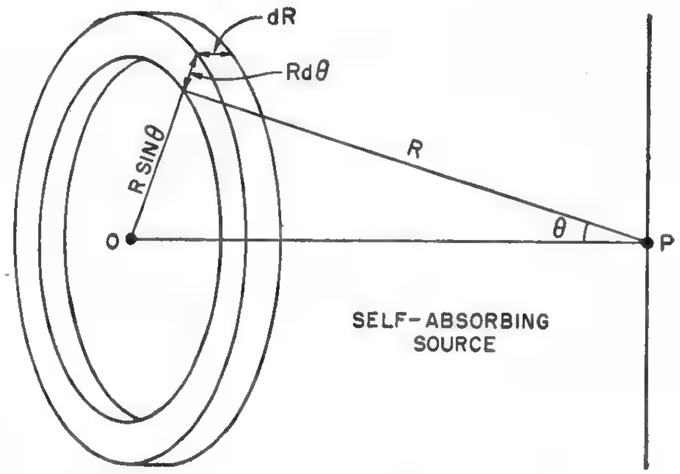


FIG. 10.7. Volume-distributed source with self-absorption

10.83. The build-up factor within the source may be taken as unity, because most of the radiation reaching the surface arises from regions near the surface that has traversed only a few relaxation lengths in the volume source. Much of the radiation from the more distant regions will be absorbed before reaching the surface. Consequently it is sufficient to set $B(R)$ equal to unity and to represent $G(R)$ by $e^{-R/\lambda}/4\pi R^2$; the integral in equation (10.82.1) can then be readily evaluated. The result is

$$\text{Radiation flux at surface} = \tfrac{1}{2} S_v \lambda, \quad (10.83.1)$$

where λ is the relaxation length for the given radiation within the volume source.

10.84. In order to make use of this result, in conjunction with the equations derived previously, such as equation (10.60.3), to calculate the dose rate at a distance z from the surface of the volume-distributed source, it must be recalled that the equations assume an isotropic source, i.e., with particles emitted uniformly in all directions. The radiation flux given by equation (10.83.1) refers to particles leaving the surface in the outward direction only and so it is equivalent to an isotropic surface source with a strength equal to twice this value.

Consequently the strength, S_a, of an isotropic surface source which is equivalent to the volume source is given by

$$S_a = S_v \lambda. \quad (10.84.1)$$

It may be noted that since S_v is expressed as particles/(cm³)(sec) and λ is usually given in cm, S_a will have the correct dimensions of a surface source strength, namely, particles/(cm²)(sec).

10.85. Although the result represented by equation (10.84.1) has been derived here for the special case of an infinitely large volume source and a build-up factor of unity, it can be shown that it is a good approximation in all cases, including systems of finite size. The relationship may therefore be used to calculate the strength of an isotropic surface source which is roughly equivalent to a given volume-distributed source. It is then possible to determine the radiation dose rate after passage through a specified shield.

Example Suppose the core of a natural uranium-graphite reactor, such as the X-10 reactor at Oak Ridge, consists of a 20-ft cube, containing roughly 50 tons of uranium and 400 tons of graphite. Estimate the gamma-ray strength in terms of Mev/(cm²)(sec) of an equivalent isotropic source at the surface of the reactor when it operates at a power of 3800 kw.

A power output of 1 watt requires 3.1×10^{10} fissions/sec (§ 1.86) and so in the given reactor the fission rate is $(3.1 \times 10^{10})(3800 \times 10^3) = 1.2 \times 10^{17}$ fissions/sec. The volume of the core is $(20 \times 30.5)^3 = 2.3 \times 10^8$ cm³, so that

$$\text{Fissions/(cm}^3\text{)(sec)} = \frac{1.2 \times 10^{17}}{2.3 \times 10^8} = 5.2 \times 10^8.$$

The total energy from both prompt (instantaneous) gamma rays and gamma radiation accompanying radioactive decay of the fission products is 12 Mev/fission. In addition about another 8 Mev are liberated, per fission, in the form of capture and decay gamma radiation within the reactor (see Table 1.5). Consequently some 20 Mev of gamma radiation energy are produced per fission. The volume source strength may therefore be expressed as

$$S_v = (20)(5.2 \times 10^8) = 1.04 \times 10^{10} \text{ Mev/(cm}^3\text{)(sec)}.$$

The next problem is to determine the relaxation length, which is equal to $1/\mu$, where μ cm⁻¹ is the linear absorption coefficient of the gamma rays produced in the reactor core. The exact distribution of the energy in these radiations is very complex, but it appears to be a satisfactory compromise to suppose that it has the absorption properties of 4-Mev gamma rays. The required value of μ may be estimated in several ways of which perhaps the simplest is the following. From the masses of uranium and graphite in the core and the known volume, the average density of the core is found to be 1.76 g/cm³. According to the data in Table 2.5 (see also § 10.94), the mass absorption coefficient, μ/ρ, for many substances is about 0.032 cm²/g for 4-Mev gamma rays. In the present case, therefore, since ρ is 1.76 g/cm³, the required value of μ is about 0.056 cm⁻¹ and the relaxation length is $1/0.056$ cm⁻¹ = 18 cm. It follows then, from equation (10.84.1), that the required equivalent isotropic surface source strength is

$$S_a = (1.04 \times 10^{10})(18) = 1.9 \times 10^{11} \text{ Mev/(cm}^2\text{)(sec)}.$$

10.86. Attention should be called to the fact that equation (10.84.1) applies only to a volume-distributed source of uniform strength, as postulated in § 10.82.

A reactor, as a general rule, is, however, not a uniform source, so that the result obtained is not strictly applicable. If the source strength (or power) variation, S_v, can be represented by a simple expression, e.g., a cosine function, as is often the case, it may be possible to perform the integration of equation (10.82.1) analytically and obtain a result for the equivalent surface source strength. A more approximate approach is to use an average value for the source strength and assume it to be constant throughout the reactor core. This method tends to overestimate the strength of the equivalent surface source, since it gives equal weight to all parts of the volume-distributed source, whereas, as indicated earlier, in actual fact most of the radiation reaching the surface arises from the outer regions of the volume where the source strength is less than in the interior.

SHIELDING OF GAMMA-RAY SOURCES

Calculation of Source Strength

10.87. As stated in § 10.63, the results obtained in the preceding sections can be used to determine the effect of a shield in attenuating either neutrons or gamma radiations. The presence of neutrons introduces complications, mainly due to production of secondary gamma rays which must be taken into consideration, but for pure gamma-ray sources, such as pipes, vessels, and pumps used for radioactive liquids, the shielding problem is very much simpler. It is quite satisfactory in such cases to design the shielding on the basis of straightforward calculations using known absorption coefficients of the proposed shielding materials for the particular gamma radiation which is to be absorbed.

10.88. If a vessel contains g grams of a radioactive species with mass number A and half life $t_{\frac{1}{2}}$ sec, the total rate of decay is given by equation (2.15.2) as $4.17 \times 10^{23} g/At_{\frac{1}{2}}$ dis/sec. If γ is the average number of gamma-ray photons produced in each act of radioactive decay (cf. § 9.50), then

$$\text{Rate of gamma-ray emission} = \frac{4.17 \times 10^{23} g\gamma}{At_{\frac{1}{2}}} \text{ photons/sec.} \quad (10.88.1)$$

When a mixture of radioelements is present, allowance must be made for all the gamma emitters in the vessel. In general, it is preferable to express the rate of emission in terms of energy. Equation (10.88.1) can then be written in the form

$$\text{Rate of gamma-ray energy emission} = 4.17 \times 10^{23} \sum \frac{gE\gamma}{At_{\frac{1}{2}}} \text{ Mev/sec,} \quad (10.88.2)$$

where E is the photon energy in Mev; the summation is carried over all the active species present. If the result is divided by the volume of the vessel, there will be obtained the source strength, expressed in Mev/(cm³)(sec), of the uniform volume-distributed source of gamma radiation. From this the corresponding

equivalent surface source strength can be calculated and the thickness of shielding required to reduce the radiation to a prescribed value can then be determined.

10.89. In the case just cited it is assumed that the weights of the various radioelements in the vessel to be shielded are known. However, in many instances it may be that the numbers of curies (or millicuries) present, rather than the weights, have been determined. In this event, the calculations will be based on the definition (§ 2.14) that the curie represents a disintegration rate of 3.7×10^{10} per sec. The expression for the rate of gamma-ray energy emission then becomes

$$\text{Rate of gamma-ray energy emission} = 3.7 \times 10^{10} \sum CE\gamma \text{ Mev/sec,}$$

where C is the number of curies of radioelement present. The source strength can now be evaluated in the manner already described.

10.90. Although these two examples cover many cases encountered in pure gamma-ray shielding, other circumstances arise from time to time. One of these is concerned with the activity induced in a coolant, as a result of neutron absorption, during its passage through a reactor. This situation will be considered shortly.

Gamma-ray Absorption Coefficients

10.91. In order to complete the calculations of shield thickness it is necessary to know, of course, the relaxation lengths (or the linear absorption coefficients) of the given gamma radiation both in the volume source and in the shielding material. Considerable data, some of which were given in Chapter II, are available concerning absorption coefficients of various substances for gamma radiation over a large range of energies.* However, unless the radiation to be shielded against is monochromatic, i.e., it consists of photons of a single energy or has a well-defined energy spectrum, so that the energy distribution is known, the exact absorption coefficients are of little use in shielding calculations.

10.92. For most practical purposes it is adequate to employ a single value of the linear absorption coefficient for each absorbing material, namely, that corresponding to a photon energy of about 4 Mev. It happens that, for most substances of interest in shielding, this is at, or close to, the minimum to which reference was made in § 2.64. Consequently, if this value is used in calculating the shield thickness required in any particular case, the result is almost certain to be somewhat conservative. This is, of course, a desirable feature, provided a small increase in the mass of the shield can be tolerated.

10.93. Some values, suitable for shielding calculations, of the linear and mass absorption coefficients of a number of materials of interest to the reactor field are recorded in Table 10.2. Data for a few substances, such as beryllium, beryl-

* W. S. Snyder and J. L. Powell, Report AECD-2739; G. R. White, National Bureau of Standards Report 1003.

TABLE 10.2. GAMMA-RAY ABSORPTION COEFFICIENTS FOR SHIELD DESIGN

Material	μ (cm^{-1})	ρ (g/cm^3)	μ/ρ (cm^2/g)
Uranium	0.72	18.7	0.038
Tungsten	0.68	19.3	0.035
Lead	0.48	11.3	0.042
Iron	0.27	7.8	0.034
Beryllium oxide	0.076	2.3	0.033
Concrete	0.073	2.3	0.032
Boron carbide	0.072	2.5	0.029
Beryllium	0.053	1.85	0.029
Graphite	0.052	1.62	0.032
Water	0.033	1.00	0.033
Sodium	0.030	0.93	0.032

lium oxide, graphite, and sodium, which would not normally be used in shields, are included because they may be present in a reactor and thus make an incidental contribution to the shielding.

10.94. If the radiation absorption is exponential in character, with a build-up factor that is either unity or linear, the thickness of material required to produce a specified fraction of absorption is inversely proportional to the linear absorption coefficient. In view of the parallelism between the values of μ and of the densities ρ in Table 10.2, it is evident that the more dense the material, the thinner the shield. The relative mass of shielding material, for a given degree of attenuation, is equal to the thickness multiplied by the density and hence is inversely related to μ/ρ, the mass absorption coefficient (§ 2.66). Values of the latter, as recorded in the last column of Table 10.2, are seen to be approximately constant, so that the weight of the gamma-ray shield is essentially independent of the material. As pointed out in § 2.67, this approximate constancy of μ/ρ, at about 0.032 cm^2/g, may be used to obtain a rough value of the linear absorption coefficient for 4-Mev gamma radiation of any material of known density.

Calculation of Shield Thickness

10.95. With the information given above and some of the results derived in Chapter IX, it is now a relatively simple matter to calculate the thickness of shield required to reduce the radiation dose rate to any specified value. The general procedure to be used for a point source was given in Chapter IX, and the somewhat more complicated problem of a volume-distributed source will be considered here. It was seen in § 9.61 that the minimum permissible dose rate of 7.5 mr/hr corresponds to a gamma-ray energy flux of about 4000 Mev/(cm^2)(sec), assuming an exposure of 40 hr/week. To determine the thickness of a shield which will reduce the radiation to a value not exceeding the maximum permissible amount, it is necessary to relate this intensity to the energy source strength, as given by equation (10.88.2), using equations already derived. The general procedure may be illustrated by means of an example.

Example As it circulates, the liquid sodium coolant of a reactor, operating at a flux of 4×10^{13} thermal neutrons/(cm^2)(sec), passes through a horizontal cylindrical tank, 4 ft long and 2 ft in diameter. Determine the thickness of a shield of special concrete (density 3.4 g/cm^3) required to reduce the gamma-ray dose from the end of the tank (due to sodium-24) to the maximum permissible value. Assume that about half of the circulating sodium is within the reactor itself, and that the cycle time is short in comparison with the half life of sodium-24 (15 hr).

If all the sodium were exposed to neutrons, the equilibrium activity of the sodium-24 would be $\Sigma\phi$ dis/(cm^3)(sec), where Σ is the macroscopic absorption cross of sodium for thermal neutrons and ϕ is the flux, in the usual units (cf. § 2.121). However, in the present case only half of the sodium atoms present are exposed to neutrons in the reactor, but all the sodium-24 formed decays. It is found, therefore, that the equilibrium activity of the sodium-24 is $\frac{1}{2}\Sigma\phi$ dis/(cm^3)(sec) within the reactor. It has been postulated that the time the sodium spends outside the reactor is short in comparison with the half life of sodium-24; consequently the activity of the sodium in the tank (outside the reactor) may be taken as equal to the equilibrium activity in the reactor.

The microscopic absorption cross section for sodium is 0.46 barn; its atomic weight is 23 and the density of the liquid is about 0.93 g/cm^3. It follows from equation (2.113.2), therefore, that Σ is 0.011 cm^{-1}. Since ϕ is 4×10^{13} neutrons/(cm^2)(sec), it is found that the equilibrium activity is given by

$$\text{Activity of sodium-24} = (\tfrac{1}{2})(0.011)(4 \times 10^{13}) = 2.2 \times 10^{11} \text{ dis/(cm}^3\text{)(sec)}.$$

Every disintegration of a sodium-24 nucleus is accompanied by the emission of two photons, with energies of 1.38 and 2.75 Mev, respectively. Hence the rate of gamma-ray energy emission is obtained upon multiplying the disintegration rate by 1.38 + 2.75 = 4.13 Mev; thus

$$\text{Rate of gamma-ray energy emission} = (2.2 \times 10^{11})(4.13) = 9.1 \times 10^{11} \text{ Mev/(cm}^3\text{)(sec)}.$$

This quantity represents the actual volume source strength per unit volume, and the equivalent (isotropic) surface source strength per unit area is obtained upon multiplying by the relaxation length of the gamma rays in the sodium (cf. § 10.84). From Table 10.2 it is seen that μ is 0.030 cm^{-1} and so the required relaxation length is 33 cm. The value of S_a is thus $(33)(9.1 \times 10^{11}) = 3.0 \times 10^{13}$ Mev/(cm^2)(sec). It may be pointed out that this estimate is probably rather high, because the relaxation lengths of 1.38- and 2.75-Mev gamma rays, for self-absorption in sodium, are certainly less than 33 cm. However, since it is usually preferable to be on the safe side in shielding calculations, the result derived above will be used.

By combining equation (10.74.1) for the dose rate from an infinite plane source, assuming a linear build-up factor, with the conversion factor $\frac{1}{2} + \alpha$ (§ 10.78), it is found that for a disk source of finite size

$$D \approx \frac{\frac{1}{2}S_a\, e^{-z/\lambda}}{\frac{1}{2} + \frac{2\lambda^2}{a^2}\left(\frac{z}{\lambda} + 1\right)}.$$

In the present case the finite disk is the end of the cylindrical tank, so that a is 1 ft, i.e., 30.5 cm; S_a is the equivalent surface source strength derived above, i.e., 3.0×10^{13} Mev/(cm^2)(sec); and λ is the relaxation length, equal to $1/\mu$, of the gamma radiation in the specified concrete. If the latter is not known, it may be estimated from the

conclusion reached in § 10.94, namely, that μ/ρ has the approximately constant value of 0.032 cm²/g for gamma rays of interest in shielding. The density of the concrete is given as 3.4 g/cm³, so that μ is about 0.11 cm^{-1} and λ is, consequently, 9.0 cm.

In order to determine the thickness, z, of shield required to reduce the radiation to the maximum permissible value, D in the equation given above is set equal to 4000 Mev/(cm²)(sec), these being the correct units, and the values for S_a, a, and λ are inserted; thus

$$4000 = \frac{(\frac{1}{2})(3.0 \times 10^{13})\, e^{-z/9}}{\frac{1}{2} + \frac{(2)(9)^2}{(30.5)^2}\left(\frac{z}{9} + 1\right)}.$$

This transcendental equation may be solved by trial-and-error or graphically; the solution is found to be $z = 190$ cm (approximately) or 6.3 ft, which is the required shield thickness.*

10.96. It should be noted that the result just obtained represents a maximum value of the shielding required, for several reasons, only two of which need be mentioned here. In the first place, the calculations are based on the supposition that the detector is located along the axis of the cylinder, which is the point of closest approach. Second, the tacit assumption has been made that there is no gap between the end of the tank and the face of the shield. In actual practice, there will almost always be an appreciable space, and this will result in purely geometric attenuation according to the inverse-square law. The amount whereby the thickness of the shield can be decreased to take advantage of this attenuation can be readily calculated (cf. § 10.107).

10.97. There is another matter in connection with the foregoing calculation to which attention should be called. It is the fact that the length of the cylinder containing the sodium does not enter into the final result. This cannot, of course, be strictly correct, but since, as already mentioned, the more distant source regions contribute a relatively small proportion of the actual radiation dose rate at the detector, the length of the cylindrical vessel may be ignored in approximate calculations, such as those made above.

DESIGN OF REACTOR SHIELDS

Introduction

10.98. The most fundamental procedure for designing a shield for a reactor would be to make use of basic data concerning absorption and scattering cross sections of neutrons and the absorption coefficients of gamma rays together with transport theory (§ 3.8) or its equivalent, to describe the passage of neutrons and photons. However, the phenomena occurring in a reactor shield are so complex, as seen in the early sections of this chapter, that this approach would be extremely difficult. In addition, the detailed knowledge of cross sections for elastic and inelastic scattering of neutrons over a considerable range of energies,

* If the infinite-plane to finite-plane correction factor ($\frac{1}{2} + \alpha$) is omitted, solution of the equation is greatly simplified; z is then found to be 200 cm, which is a good first approximation.

which would be necessary for these calculations, is not available. It is possible that in due course reactor shields will be designed in this manner, but at the present time recourse must be had to other procedures which are either approximate or are largely empirical in character. These procedures fall into two general categories, namely, (1) direct calculation methods which utilize relaxation lengths or cross sections and (2) comparison methods based on experimental measurements with shield mock-ups.

Use of Relaxation Lengths

10.99. Shields for stationary reactors, when weight is not of prime significance, are designed with a considerable factor of safety, so that approximate calculations are adequate to provide at least preliminary design information. Such calculations, which are similar to those already described in connection with the shielding of gamma-ray sources, make use of known (or estimated) relaxation lengths for fast neutrons and gamma rays. Some approximate values of these relaxation lengths for materials of interest in shielding are recorded in Table 10.3. Data for graphite, beryllium, and beryllium oxide are included, since the

TABLE 10.3. RELAXATION LENGTHS FOR FAST NEUTRONS AND GAMMA RAYS

Material	Density (g/cm³)	Relaxation Length (cm)	
		Fast Neutrons	Gamma Rays
Water	1.0	~10	30
Graphite	1.62	~9	19
Beryllium	1.85	~9	18
Beryllium oxide	2.3	~9	14
Concrete	2.3	11	15
Aluminum	2.7	~10	13
Barytes concrete	3.5	8.0	10
Iron concrete	4.3	6.3	8
Iron	7.8	~6	3.7
Lead	11.3	~9	2.5

reflector may make an appreciable contribution to the attenuation of the radiation escaping from a reactor core.

10.100. It should be understood that the figures in Table 10.3 represent something in the nature of a compromise. Both the neutrons and the gamma radiation from a reactor cover a considerable energy spectrum, so that in exact shielding calculations it would be necessary to take into account the nature of these spectra and the variation of the relaxation lengths with energy. However, because of the complexity of the situation, it is doubtful whether this can yet be done with any degree of certainty. Until more precise (but more involved)

methods are developed, it is probably sufficient to use a simple approach based on a single, partly empirical, relaxation length for each type of reactor radiation.

10.101. It will be apparent, from the fact that no data have been included for thermal neutrons, that such neutrons are neglected in shielding calculations. It has been found by experience, in agreement with expectation, that, provided a suitable shielding material is used, as explained earlier, the absorption of slow neutrons is no problem. Any shield which sufficiently attenuates the fast neutrons and the primary and secondary gamma radiations will inevitably reduce the thermal neutrons to an adequate extent.

10.102. The calculation of the shield thickness required to decrease the fast neutrons and primary gamma rays to permissible values involves the determination, first, of the volume source strength per unit volume. This is then converted into the equivalent (isotropic) surface source strength per unit area, and the shield thickness necessary to produce a prescribed dose rate is then calculated in the manner already described for gamma rays.

10.103. The treatment of the secondary gamma rays, which are formed as a result of inelastic scattering and ultimate capture of the fast neutrons within the shield itself, however, presents a difficulty. Since these radiations are produced more or less throughout the whole thickness of the shield, it is not possible to ascribe to them a definite source. An approximate method is consequently used which is known to represent a conservative (or safe) approach to the problem. In this treatment it is supposed that all the fast neutrons entering the (biological) shield are captured at a distance within the shield equal to τ/λ, called the *age displacement*, where τ is the Fermi age (§ 3.90) of thermal neutrons in the shield material and λ is the relaxation length for fast neutrons. In other words, the secondary gamma rays are all supposed to emanate from a plane lying a distance τ/λ within the shield (Fig. 10.8). Since the value of τ for the shield material is not always known, it is probably a close enough approximation to assume that in most cases the age displacement is about 30 cm or 1 ft.

10.104. The fluxes corresponding to the maximum permissible radiation exposure, as given in Chapter IX, are about 30 fast neutrons/(cm²)(sec) or 4000 Mev photons/(cm²)(sec). Since the radiation escaping from a reactor will consist of both fast neutrons and photons, the permissible amounts, used in calculating shield thickness, must be less than these values. For purposes of illustration, in the example given below, it will be supposed that the neutron dose rate must not exceed 10 fast neutrons/(cm²)(sec) and that the primary and secondary gamma-ray energy fluxes must each be not more than 1000 Mev/(cm²)(sec), so that the total dose would still be somewhat less than the maximum permissible amount.

Example A reactor of the Oak Ridge X-10 (natural uranium) type consists of a 20-ft cube core surrounded by a 2-ft thick graphite shield, as described in the example in § 10.85. Estimate the thickness of an ordinary concrete shield that would be required for such a reactor, for operation at a power of 3800 kw.

Fast Neutrons As seen in the example cited, when operating at the specified power, fissions occur in the reactor at the rate of $5.2 \times 10^8/(\text{cm}^3)(\text{sec})$. Since each act of fission of uranium-235 is accompanied by the liberation of about 2.5 fast (fission) neutrons (Table 1.3), the fast-neutron (volume) source strength may be taken as

$$\begin{aligned}\text{Fast-neutron source strength} &= (2.5)(5.2 \times 10^8) \\ &= 1.3 \times 10^9 \text{ fast neutrons}/(\text{cm}^3)(\text{sec}).\end{aligned}$$

The equivalent surface source strength is then obtained, in accordance with equation (10.84.1), upon multiplying this result by the relaxation length of fast neutrons in the

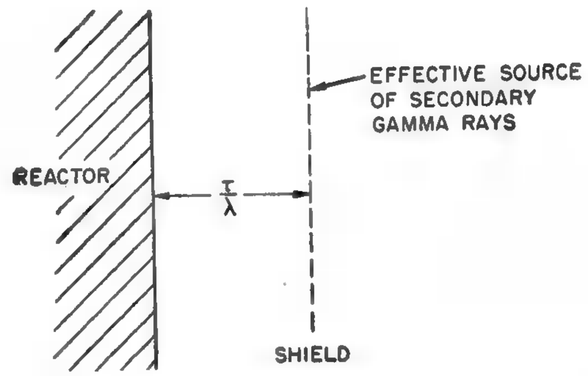


FIG. 10.8. Effective source of secondary gamma radiation from reactor

reactor core. Since the latter consists mainly of graphite, the required relaxation length may be taken as 9 cm (Table 10.3), although this is probably somewhat of an overestimate. Consequently,

$$\begin{aligned}\text{Equivalent surface source strength} &= (1.3 \times 10^9)(9) \\ &= 1.2 \times 10^{10} \text{ fast neutrons}/(\text{cm}^2)(\text{sec}).\end{aligned}$$

The next step is to determine the attenuation of the fast neutrons in the 2-ft thick graphite reflector. This can be obtained by means of equation (10.74.1) with S_a equal to $1.2 \times 10^{10}/(\text{cm}^2)(\text{sec})$, λ being 9 cm, and z is 61 cm (2 ft); thus

$$\begin{aligned}\text{Fast neutrons leaving the reflector} &= \frac{1.2 \times 10^{10}}{2} e^{-61/9} \\ &= 6.6 \times 10^6 \text{ fast neutrons}/(\text{cm}^2)(\text{sec}).\end{aligned}$$

This quantity now represents half the isotropic surface source term for the concrete shield (cf. § 10.84); for the latter, the relaxation length for fast neutrons is given in Table 10.3 as 11 cm, and since the permitted dose has been set at 10 fast neutrons/(cm²)(sec), the thickness z of the shield is given by

$$10 = \frac{(2)(6.6 \times 10^6)}{2} e^{-z/11},$$

so that z is readily found to be about 150 cm.

Primary Gamma Rays This aspect of the problem is exactly the same as that considered in the example following § 10.85. It was shown there that the equivalent sur-

face source strength, in terms of the energy flux, is roughly 1.9×10^{11} Mev/(cm²)(sec). Taking the relaxation length for the primary gamma rays in graphite as 19 cm, the gamma-ray energy flux escaping from the reflector is found to be

$$\text{Gamma-ray energy leaving the reflector} = \frac{1.9 \times 10^{11}}{2} e^{-61/19}$$
$$= 3.8 \times 10^{9} \text{ Mev/(cm}^2\text{)(sec)}.$$

and the isotropic source term for the concrete shield is twice this amount. The permissible dose rate for the primary gamma rays is to be taken as 1000 Mev/(cm²)(sec), and since the relaxation length for gamma rays in ordinary concrete is 15 cm, the required shield thickness is obtained by solving the equation

$$1000 = \frac{(2)(3.8 \times 10^{9})}{2} e^{-z/15},$$

from which z is 230 cm.

Secondary Gamma Rays As seen above, the equivalent (isotropic) fast-neutron surface source strength leaving the reflector and entering the concrete shield is $(2)(6.6 \times 10^{6})$ neutrons/(cm²)(sec). Each neutron may be supposed to produce one secondary gamma-ray photon of 4-Mev energy, originating at 30 cm within the shield, as postulated in § 10.103. The thickness of shield required to attenuate these secondary gamma rays to 1000 Mev/(cm²)(sec), is then obtained from the relationship

$$1000 = \frac{(4)(2)(6.6 \times 10^{6})}{2} e^{-z/15},$$

according to which z is 150 cm. To this must be added the 30 cm allowance for the origin of the radiation, so that the total shield thickness required would be 150 + 30 = 180 cm.

It is evident in the present case that the limiting factor in determining the shield thickness is the primary gamma radiation; this requires a thickness of about 230 cm of concrete. Such a shield will reduce the fast neutrons and the secondary gamma rays to negligible proportions, and the primary gamma rays will produce a dose rate roughly one fourth of the maximum permissible value.

(The shield of the X-10 reactor consists of 2 ft of ordinary concrete and 5 ft of a special concrete of density 2.4, so that the total would be equivalent to roughly 7.23 ft, i.e., about 220 cm, of ordinary concrete. The agreement with the calculated thickness of 230 cm is probably as good as could be expected, since single (averaged) values were used for the relaxation lengths and all data were restricted to two significant figures. It would appear, therefore, that the procedure described may be employed, as stated earlier, to provide a useful first approximation for shield design.)

10.105. In the example given above the absorption of the primary gamma radiation determined the shield thickness. It happens that for this particular reactor the fast neutrons and gamma rays must each be attenuated by a factor of a little over 10^{-8}, and since the relaxation length in both graphite and concrete is greater for gamma rays than for neutrons (Table 10.3) the former require a greater thickness of shield. If the shielding material had contained a large proportion of iron, lead, or other heavy element the situation could very well have been reversed, since in these cases the relaxation length is larger for fast neutrons than it is for gamma rays. Another possibility is that the reactor characteristics

are such that the fast neutrons need to be attenuated to a much greater extent than the primary gamma radiation. In this event, the fast neutrons, or even the secondary gamma rays, might determine the shield thickness.

10.106. It may be noted, too, that according to the calculations given the X-10 shield does not by any means satisfy the requirement for an efficient shield mentioned in § 10.30, namely, that the fast neutrons and gamma rays should be reduced to their maximum permissible values at about the same place. This is, of course, again due to the nature of the shielding material, in which the relaxation length is appreciably greater for gamma rays than for fast neutrons, although both radiations have to be attenuated by roughly the same factor. Some improvement would be achieved by the use of a heavy concrete, although more nearly ideal conditions would result if sheets or layers of iron or lead could be introduced into the shield in some manner. An efficient shield, from the standpoint of weight, could thus be constructed from alternate layers of heavy metal and water, and such shields have, in fact, attracted some interest (§ 10.30).

Attenuation by Distance in Shield Design

10.107. If the circumstances are such that personnel can be kept at a good distance from a reactor when it is in operation, it is possible to effect a considerable saving in the thickness (and cost) of the shield. This fact should be borne in mind, since it may be highly significant in shield design. The situation arises because of the inverse-square-law attenuating effect of distance. Suppose that just outside a shield of 7 ft thickness the total radiation dose rate is equal to the maximum permissible value. At a farther distance of, say, 28 ft in air, representing a total distance of 35 ft from the reactor face, the radiation dose will be reduced to a factor of $(7/35)^2$, i.e., 0.04, of the maximum, irrespective of absorption in the air. Consequently, if it were possible to prevent operating personnel from coming closer than 28 ft from the shield exterior when the reactor was operating, it would be possible to decrease the shield thickness and still retain the same radiation dose rate.

Example By how much could the thickness of the X-10 (natural uranium) reactor shield be decreased on a face to which access was restricted to a distance of more than 35 ft from the reactor itself?

Since the thickness of the X-10 reactor shield is 7 ft, the dose rate at 35 ft from the reactor is, as seen above, 0.04 of the value just outside the shield. The factor of 0.04 is very close to $e^{-3.2}$, so that the attenuation due to distance is equivalent to 3.2 relaxation lengths. As seen in the problem given above, the primary gamma rays determine the thickness of the X-10 shield, and for these radiations the relaxation length is about 15 cm. The shield thickness could thus be reduced by about (3.2)(15) cm, i.e., 48 cm or 1.6 ft.

10.108. It is opportune to mention here that reactors must be shielded in all directions even if there is no access to one or other of the faces. For example, suppose access to the top of a reactor is completely prohibited while the reactor is in operation. Nevertheless some shielding of the top is necessary. This is

because neutrons and gamma rays escaping from the top of the reactor would be scattered downward by the nuclei (or atoms) present in the air and by the roof and walls of the building. As a result operating personnel might be exposed to radiation which reached them in an indirect manner. Some shielding is thus necessary for inaccessible faces of a reactor and, in fact, for any source of radiations, whether they are neutrons or gamma rays or both. Methods for calculating the extent of scattering have been developed, but they lie outside the scope of this book.

Effective Removal Cross Sections

10.109. In another calculation method of shield design, an attempt is made to utilize cross sections to estimate neutron attenuation. It has been applied hitherto only to shields in which water is the moderator and the main absorber of slow neutrons. Instead of using basic data, which are, in any case, not available and are difficult to apply (§ 10.98), a semiempirical approach is employed. A quantity known as the *effective removal cross section* of a material, such as lead or iron, is determined by experimental observations of the decrease in fast-neutron dose rate resulting from the insertion of a slab of this material in a water shield.

10.110. The effective removal cross section, which is partly due to scattering and partly to absorption, does not have a precise physical significance, since it may be expected to vary with neutron energy and, consequently, with the position of the material in the shield. Nevertheless, empirical removal cross sections, assumed to be independent of energy, have been assigned to various materials. It appears, from theoretical considerations, that, for elements of low mass number, the effective removal cross section should be almost equal to the total cross section for interaction with fast neutrons, i.e., to $2\pi R^2$, where R is the nuclear radius (cf. § 10.18), since almost every interaction of a fast neutron with a nucleus results in the ultimate removal of the former. For heavy nuclei, on the other hand, only about half of the elastic collisions are effective in causing removal of fast neutrons, although essentially all of the inelastic collisions, as well as other interactions, lead to neutron removal. The effective removal cross sections are then roughly equal to $\frac{2}{3}\pi R^2$. For nuclei of intermediate mass the effective removal cross sections lie between the two values indicated.

10.111. Suppose a shield consists of a z cm layer of water and a slab of thickness x cm of another material which either scatters or absorbs fast neutrons, or does both. Let $D_0(z)$ be the dose rate observed from a given source (or reactor) after passing through a thickness z cm of water only. Then the dose rate $D(z, x)$ from the same source for the postulated shield will be

$$D(z, x) = e^{-\Sigma_r x} D_0(z), \qquad (10.111.1)$$

where Σ_r cm^{-1} is the effective macroscopic removal cross section of the material;

this is related to the effective (microscopic) removal cross section, σ_r, in the usual manner, by equation (2.113.2).

10.112. Some values for the effective removal cross sections of a number of elementary materials in a water shield are recorded in Table 10.4. These results can be fairly well represented over a considerable range of atomic weights by the expression

$$\sigma_r = 0.011A^{\frac{2}{3}} + 0.56A^{\frac{1}{3}} - 0.35 \text{ barn},$$

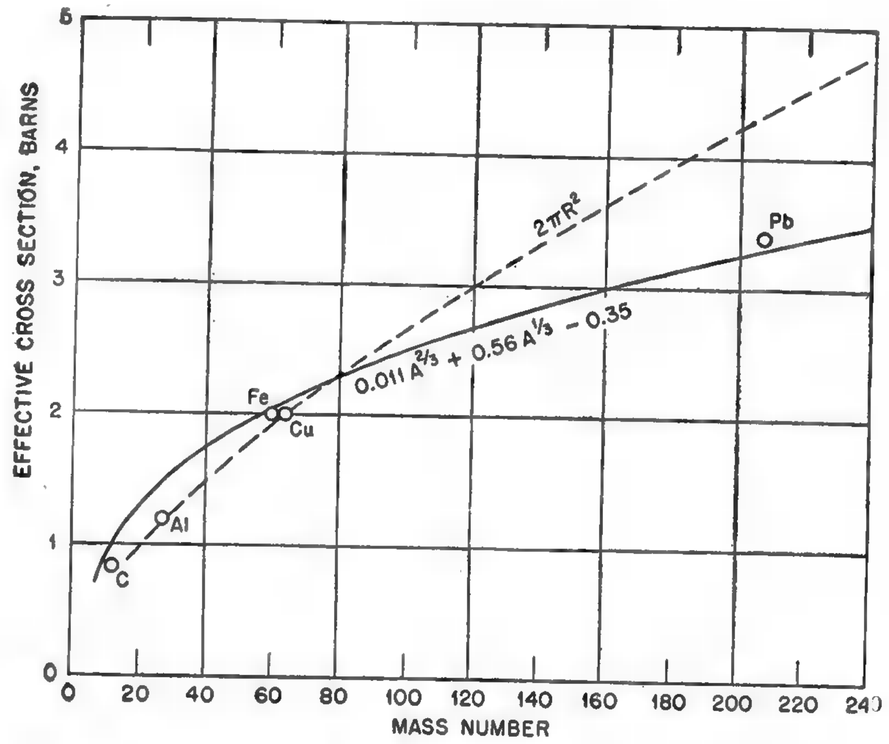


Fig. 10.9. Effective removal cross sections for fast neutrons (E. P. Blizard)

where A is the atomic weight of the element. This is shown by Fig. 10.9 in which the full line is a plot of this equation, with the experimental points inserted for a number of cases. As a matter of interest, the dotted curve indicates the

Table 10.4. Effective Removal Cross Sections for Fast Neutrons

Material	*At. Wt.*	σ_r, barns
Lead	207	3.4
Iron	56	2.0
Copper	63.5	2.0
Aluminum	27	1.2
Carbon (graphite)	12	0.84

values of $2\pi R^2$ (n barns), which may be taken as the total cross section for fast-neutron interaction.

10.113. In principle, the data in Table 10.4 might be used in conjunction with equation (10.11[illegible].1) to calculate the approximate dose rate of fast neutrons that would penetrate any shield consisting of x cm of the material and z cm of water surrounding a specified reactor. The other information necessary would be the fast-neutron dose rate for the same reactor with a shield of z cm of water only. This can be obtained from experimental measurements.

10.114. Rather than place too much reliance on the effective removal cross sections, as would be required by the procedure in the preceding paragraph, a preferable application would be to use the data to determine the effect of replacing one materia by another in a particular water shield. The calculations are based on the requirement of the exponential absorption law that the thickness of material needed to produce a certain degree of attenuation of radiation is inversely proportional to the macroscopic cross section.

Example Compare the thicknesses and masses of iron and lead which would be required to produce a specified degree of attenuation of fast neutrons in a given water shield.

According to the exponential absorption law, e.g., equation (10.111.1), a specified degree of attenuation of fast neutrons means that $\Sigma_r x$ is constant, and so the thickness, x cm, of iron or lead required is inversely proportional to its macroscopic effective removal cross section. According to equation (2.113.2),

$$\Sigma_r \text{ for iron} = \frac{(7.8)(N_a)}{(56)} 2.0 \times 10^{-24} \text{ cm}^{-1},$$

since the density of iron is 7.8 g/cm³ and its atomic weight is 56; σ_r, as given in Table 10.4, is 2.0 barns. Similarly

$$\Sigma_r \text{ for lead} = \frac{(11.3)(N_a)}{(207)} 3.4 \times 10^{-24} \text{ cm}^{-1},$$

where the density is 11.3 g/cm³, the atomic weight is 207, and σ_r is 3.4 barns. Hence,

$$\frac{\Sigma_r \text{ for iron}}{\Sigma_r \text{ for lead}} = 1.5,$$

and the relative thicknesses, which are inversely proportional to the macroscopic cross sections, are then 1.5 of lead to 1 of iron. The relative masses are obtained upon multiplying by the ratio of the densities; the result is that a given mass of iron is as effective as 2.2 times that mass of lead, for the attenuation of fast neutrons in a given water shield. (This is in agreement with the prediction in § 10.19 that an element of lower mass number will give a neutron shield of lighter weight than one of higher mass number.)

10.115. It should be emphasized that the effective removal cross sections are strictly applicable to shields consisting of alternate layers of the material and water, so that essentially no neutrons travel very far after their first interactions before they are slowed down and captured. In general, it is necessary that the shield contains an appreciable proportion of hydrogen, fairly uniformly distributed, if the data in Table 10.4 or Fig. 10.9 are to be used for calculations. The minimum proportion of hydrogen that will satisfy the requirement is not known exactly, but it is probably met by something of the order of 10 per cent of the weight of the shield as water.

Gamma-ray Replacement Lengths

10.116. Because of the production of secondary gamma radiation throughout a large proportion of the reactor shield, the direct calculation of attenuation is very difficult and an approximate procedure was described in § 10.103. Another method of calculation is based on the use of empirical relaxation lengths somewhat analogous to the effective removal cross sections for neutrons. The effect of introducing a heavy metal, such as lead or iron, into a water shield can be expressed in terms of a *replacement length*, l. This is defined in the following manner. Let Γ_1 be the gamma-ray dose rate as received at the exterior of a shield containing water, and let Γ_2 be the value when a layer of metal of thickness t cm is introduced, replacing some of the water; then

$$\Gamma_2 = \Gamma_1 e^{-t/l}, \tag{10.116.1}$$

indicating that the replacement length is a kind of relaxation length. For practical purposes, equation (10.116.1) may be written as

$$l = \frac{t}{\ln (\Gamma_1/\Gamma_2)} = \frac{t}{2.303 \log (\Gamma_1/\Gamma_2)}, \tag{10.116.2}$$

so that l can be determined directly.

10.117. The replacement lengths are functions of many variables such as position in the shield, total shield thickness, over-all composition of the shield, and the nature of the radiation source. In spite of this they have been found to be of considerable use in estimating the effect on gamma-ray attenuation accompanying the addition or removal of small thicknesses of metal within the shield.

Example In a spherical lead-water shield, the replacement length was found to be 3.6 cm at a radius of 90 cm and 2.3 cm at a radius of 120 cm. (The value of l is less farther from the source because of the decrease in the gamma-ray energy due to scattering and the corresponding increase in the absorption coefficient.) Which is the better location of an additional layer of lead, from the standpoint of minimizing the weight of the shield?

Suppose the additional lead is to reduce the gamma-ray dose rate outside the shield by half, i.e., let $\Gamma_1/\Gamma_2 = 2$. Then, by equation (10.116.2), when r is 90 cm, so that l is 3.6 cm, t is 2.50 cm; and when r is 120 cm, l is 2.3 cm, and t is 1.59 cm. The volumes of the respective spherical shells are then $(4\pi)[90 + (\frac{1}{2} \times 2.50)]^2(2.50)$ cm³ at 90 cm, and $(4\pi)[120 + (\frac{1}{2} \times 1.59)]^2(1.59)$ cm³ at 120 cm. These quantities are in the ratio of about 1 to 1.1, so that a saving in weight would be expected if the layer of lead were placed at the 90-cm-radius position.

Comparison Method of Shield Design

10.118. There is no doubt that safe shields can be designed with the aid of calculations based on relaxation lengths or cross sections by taking a conservative approach. However, such shields may well be thicker and, consequently, heavier and more costly than is really necessary. Where weight or economic considerations are paramount, it is necessary to use the comparison method of shield design. This procedure involves, first, the proposal of a design scheme based upon a general knowledge of shielding theory and materials, such as has been presented in the earlier parts of this chapter, and upon experience. A partial mock-up of the proposed design is then constructed, and extensive measurements are made of dose rates of fast and slow neutrons and gamma radiation for various thicknesses of the shield. For this purpose a convenient radiation source, to be described below, is used, and the experimental results are transformed, by means of the equations already derived, to those to be expected from the particular geometry for which the shield is intended. An example of the calculation procedure will be given later.

10.119. Although the comparison method is the most reliable at present available for shield design, it suffers from the drawback that every fresh proposal requires the construction of a new mock-up and a repetition of the measurements. However, as information and experience are accumulated, the problem of shield design will be considerably simplified and fewer tests will be necessary.

Experimental Facilities

10.120. In recent years, much experimental work on reactor shields has been performed with the *Lid Tank Shielding Facility* (LTSF) and the *Bulk Shielding Facility* (BSF) at Oak Ridge National Laboratory. In addition, a facility somewhat similar to the LTSF, but with several improvements, has been set up at Brookhaven National Laboratory.

10.121. In the LTSF * a circular plate, 35.6 cm in radius, consisting of a closely packed, flat array of natural-uranium fuel elements, is placed over a hole which extends through one side of the concrete shield of the natural-uranium reactor at Oak Ridge (Fig. 10.10). Thermal neutrons escaping from the reactor cause the uranium-235 to undergo fission, and a large proportion of the neutrons and gamma radiation produced enter a tank of water in which the shield to be tested is inserted. The instruments used for determining the intensities of various radiations are placed in the water behind the shield.

10.122. The LTSF has a number of advantages. Many shields of interest incorporate water as an important component; such shields are very easily mocked-up in the tank. The radiation background due to leakage from the reactor itself is considerably reduced by the water, so that it is possible to measure

* This facility receives its name from the fact that it consists of a water tank which acts as a lid (or cover) to a hole through the shielding of the X-10 (natural uranium) reactor.

very low intensities of radiation, from the source plate, that penetrates the shield. The fact that the source is essentially a flat disk simplifies the transformation calculations, as will be seen below. Its main disadvantage is its relatively low source strength.

10.123. The total power output of the source has been determined from temperature measurements and found to be 6 watts. However, not all the radiations produced enter the water tank because about 40 per cent is absorbed in the walls of the box containing the source, the walls of the tank, etc.; hence the effective (isotropic) power of the source is 3.6 watts. The area is 3970 cm², and so the surface source strength may be represented as 3.6/3970, i.e., 9.1×10^{-4} watt/cm². This is too low to permit measurement of the fast-neutron intensities that have penetrated moderately thick shields.

10.124. The BSF at Oak Ridge makes use of an actual reactor as the source of radiation. The reactor core is a rectangular parallelepiped, 24 by 15 by 15 in. in size, made up of a number of fuel elements of the parallel-plate type. This is suspended in a large pool of water, as was stated in § 10.3. The reactor system, officially known as the Bulk Shielding Reactor, is sometimes referred to, for obvious reasons, as the Swimming Pool Reactor. The shields to be tested, which often have water as an important component, are placed close to the reactor, at the side or underneath.

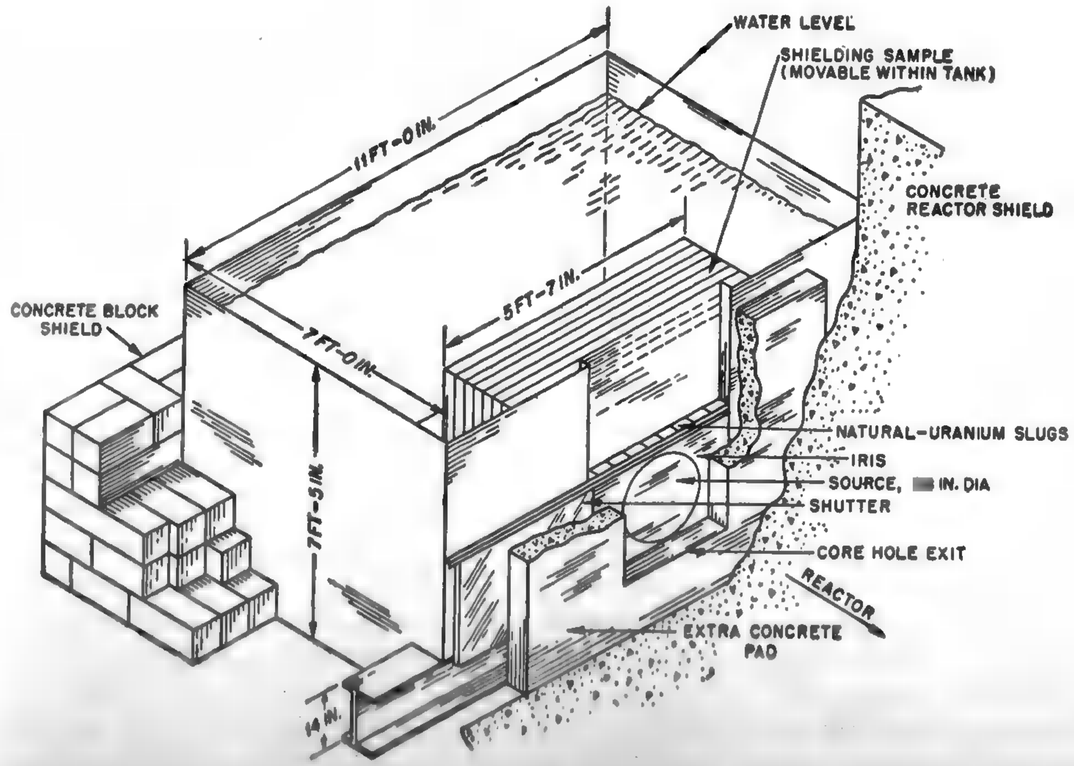


Fig. 10.10. Lid Tank Shielding Facility at Oak Ridge National Laboratory

10.125. Although the BSF lacks the simplicity of the LTSF, both as regards operation and transformation of the results, it has the great advantage of providing sufficient source strength to permit radiation intensity measurements to be made with shields of full thickness. A large range of source strengths is available, since the reactor may be operated from very low power up to about 100 kw. Further, the greater space in the BSF makes possible the mock-up of curved shields, whereas the LTSF is best adapted to measurements with slab shields.

10.126. In using any of the facilities described above for shield design by the comparison method, a mock-up of the proposed shield is constructed, and the radiation intensities which penetrate various thicknesses of the shield are determined. Measurements are made of the slow- and fast-neutron flux and of the gamma radiation by means of suitable instruments.

Application of LTSF Measurements

10.127. Data obtained with the finite disk source of the LTSF can be readily transformed to dose rates, etc., for other sources. As an illustration of the procedure employed, suppose that Fig. 10.11 represents the fast-neutron dose rates, as measured in the LTSF, for various thicknesses (z cm) of a particular type of shield. The dose rates are, for convenience, expressed in millireps per hour (mrep/hr), using the conversion factors given in Chapter IX. As stated earlier, the source in the LTSF is a disk of 35.6 cm radius and its effective source strength is 9.1×10^{-4} watt/cm².

10.128. Suppose it is required to transform the results in Fig. 10.11 so as to give the fast-neutron dose rates to be expected for the same shield when used in connection with a spherical reactor of 50 cm radius, having a power density of 1 watt/cm³. If the relaxation length of fast neutrons in the reactor core is 10 cm, the equivalent surface source strength, by equation (10.84.1), is 10 watts/cm². The problem is now to transform the dose rates obtained with a circular disk of radius 35.6 cm and source strength 9.1×10^{-4} watt/cm² to the case of a spherical surface source of radius 50 cm and strength 10 watts/cm².

10.129. Let $D_{LT}(z)$ be the dose rate observed in the LTSF at a distance z from the source plate in a specified shield. The relaxation length, λ, of fast neutrons in the shield at this point is estimated from the slope of the line in Fig. 10.11, and from this is calculated the correction factor $\frac{1}{2} + \alpha$, converting $D_{LT}(z)$ into the dose rate to be expected at z from an infinite plane reactor having the same source strength per unit area as the disk in the LTSF (§ 10.123). To obtain the dose rate at the distance z when the shield is used in connection with a spherical reactor of the same source strength, then, in accordance with equation (10.60.3), this result is multiplied by r/r_0, which in this case is $50/(50 + z)$. Finally, allowance is made for the difference in the source strengths of the LTSF and the specified spherical reactor by multiplying by the ratio of the source strengths, i.e., $10/(9.1 \times 10^{-4})$. Thus

$$D_R(z) = D_{LT}(z) \times (\tfrac{1}{2} + \alpha) \times \frac{r}{r_0} \times \frac{10}{9.1 \times 10^{-4}},$$

where $D_R(z)$ is the dose rate which would be expected from the spherical reactor with a shield z cm thick, and $D_{LT}(z)$ is the observed LTSF dose rate, in the same units, for the same shield thickness. By performing this calculation for a number of values of z, it is possible to plot a curve, equivalent to the one in Fig. 10.11, giving the dose rates for various shield thicknesses for the specified spherical reactor. The shield thickness for a particular dose rate can then be ascertained.

Example Calculate the fast-neutron dose rate to be expected from the spherical reactor referred to above (radius 50 cm, equivalent surface source strength 10 watts/cm²) for a thickness of 100 cm of shield for which Fig. 10.11 gives the dose rates in the LTSF.

For $z = 100$ cm the LTSF dose rate in Fig. 10.11 is close to 7×10^{-2} mrep/hr, and the relaxation length for fast neutrons in the shield is approximately 9 cm. (This is the distance in which the dose rate is decreased by a factor of 1/2.72 in the vicinity of $z = 100$ cm.) Then, from equation (10.78.2),

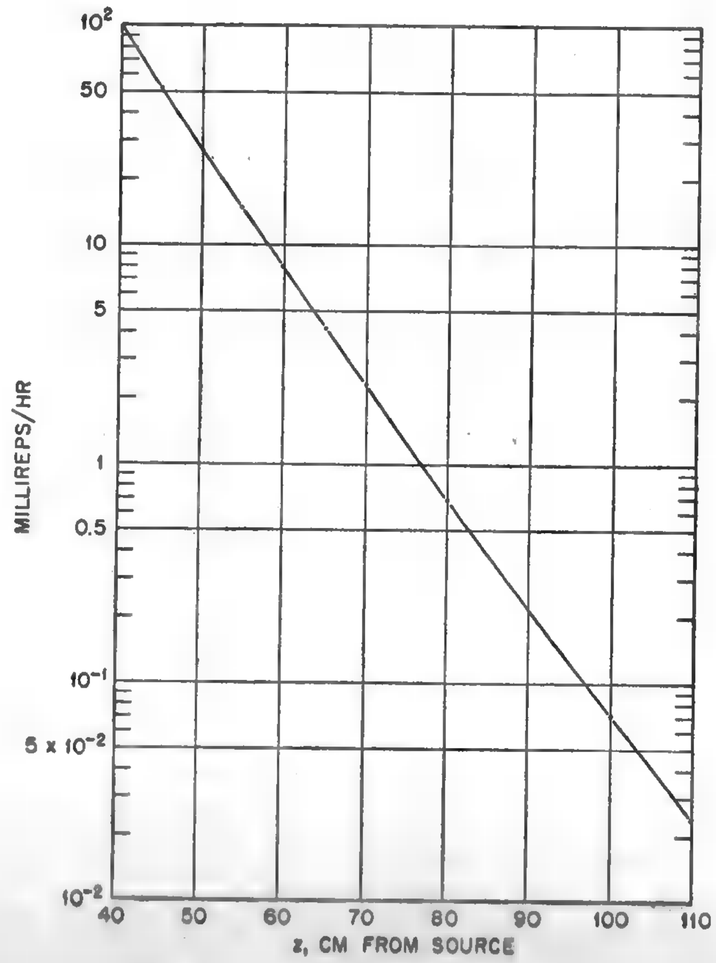


Fig. 10.11. Fast-neutron dose rates

$$\alpha = \frac{(2)(9)^2}{(35.6)^2}\left(\frac{100}{9} + 1\right) = 1.52,$$

since a, the radius of the disk source, is 35.6 cm. Consequently $\frac{1}{2} + \alpha$ is 2.02, and this is the factor for converting the LTSF dose rates to those for an infinite plane source. For a spherical source of radius 50 cm, the conversion factor for a shield thickness of 100 cm is now 50/(50 + 100), and the source strength ratio is $10/(9.1 \times 10^{-4})$. The required dose rate is then given by

$$\text{Dose rate at 100 cm from spherical reactor} = (7 \times 10^{-2})(2.02)\left(\frac{50}{150}\right)\left(\frac{10}{9.1 \times 10^{-4}}\right)$$
$$= 520 \text{ mrep/hr}.$$

(In order to reduce this to 0.5 mrep/hr, which is about two thirds of the maximum permissible dosage for fast neutrons, it can be shown that an additional 63 cm, making a total of 163 cm, of the given shield would be necessary, assuming the relaxation length remains approximately constant at 9 cm.)

Comparison Method for Gamma Rays

10.130. The foregoing discussion has referred, in particular, to the attenuation of fast neutrons because for certain shields, although not for all (cf. § 10.105), it is this radiation which determines the required thickness. However, reactor shields can be studied for their effectiveness in absorbing gamma radiation by methods analogous to those described above for fast neutrons. For gamma rays produced within the reactor, the comparison method may be applied, provided the relaxation lengths in the shield are not too long. There is some uncertainty concerning the source strengths to be used because of self-absorption of the gamma radiation within the reactor. By taking the source strength as proportional to the reactor power, as for fast neutrons, thus ignoring self-absorption, the results obtained will represent upper limits, at least.

10.131. When the gamma radiations are secondary in character, being produced mainly in the shield, as a result of various neutron interactions, the relative source strengths are the same for neutrons and for gamma rays. Thus the core relaxation length for neutrons is used in estimating the effective surface-source strength of the gamma radiation. In making the transformation from a finite disk source to an infinite plane, it is the relaxation length of the gamma rays (and not of the neutrons) in the shield that should be used, since this determines the attenuation at large distances.

APPENDIX *

This appendix contains the detailed derivations of certain results used or mentioned in earlier parts of the chapter.

* The derivations given here are based on unpublished reports by E. P. Blizard and by T. A. Welton and E. P. Blizard.

Geometrical Transformations: Planar Sources

10.132. Much of the experimental work in shielding has been carried out, in the LTSF, with a nearly isotropic circular (disk) source of finite radius (§ 10.122). Hence equations permitting transformation from such a source are very useful in shielding calculations. Consider, first, an infinite plane source; the dose rate at a distance z will be represented here by the symbol $D_{\text{pl}}(z, \infty)$, and this is given by equation (10.57.2) as

$$D_{\text{pl}}(z, \infty) = 2\pi S \int_z^\infty G(R)R\, dR.$$

Upon differentiating with respect to z, it is found that

$$\frac{d}{dz} D_{\text{pl}}(z, \infty) = -2\pi S z G(z)$$

or

$$G(z) = -\frac{1}{2\pi S z} \cdot \frac{d}{dz} D_{\text{pl}}(z, \infty). \qquad (10.132.1)$$

The point kernel, $G(z)$, can thus be determined, provided $D_{\text{pl}}(z, \infty)$ is known for several values of z, so that the derivative can be evaluated. As seen above, the point kernel is fundamental to radiation dose-rate calculations; consequently the data obtained for an infinite plane source can be utilized to treat sources of other shapes.

10.133. For a disk source of finite radius a, the dose rate at a distance z, represented by $D_{\text{pl}}(z, a)$, is given by equation (10.57.1); thus

$$D_{\text{pl}}(z, a) = 2\pi S \int_z^{\sqrt{z^2+a^2}} G(R)R\, dR.$$

If the integral is now expressed as the difference of two integrals, in which the lower limits are z and $\sqrt{z^2 + a^2}$, respectively, and the upper limits are infinity in each case, it is readily seen with the aid of equation (10.132.1) that, upon differentiation with respect to z,

$$\frac{1}{2\pi S z} \cdot \frac{d}{dz} D_{\text{pl}}(z, a) = G(\sqrt{z^2 + a^2}) - G(z), \qquad (10.133.1)$$

where $G(\sqrt{z^2 + a^2})$ and $G(z)$ are the values of the point kernels for source-to-detector separations of $\sqrt{z^2 + a^2}$ and z, respectively.

10.134. If a function $B(z)$ is defined by

$$B(z) \equiv -\frac{1}{2\pi S z} \cdot \frac{d}{dz} D_{\text{pl}}(z, a), \qquad (10.134.1)$$

then it is found from the recursion formula of equation (10.133.1) that

$$G(z) = B(z) + B(\sqrt{z^2 + a^2}) + B(\sqrt{z^2 + 2a^2}) + \cdots$$
$$= \sum_{\nu=0}^{\infty} B(\sqrt{z^2 + \nu a^2}), \qquad (10.134.2)$$

where ν is zero or integral. This means that, if the function $B(z)$ is determined for a series of z values, i.e., distances from disk source to detector, equal to z, $\sqrt{z^2 + a^2}$, $\sqrt{z^2 + 2a^2}$, $\sqrt{z^2 + 3a^2}$, etc., the sum will give $G(z)$, the point kernel at z. For this procedure to be useful, the series must converge rapidly; such is the case when the disk radius a is large compared with the relaxation length of the radiation. If a is small, then the disk is essentially equivalent to a point

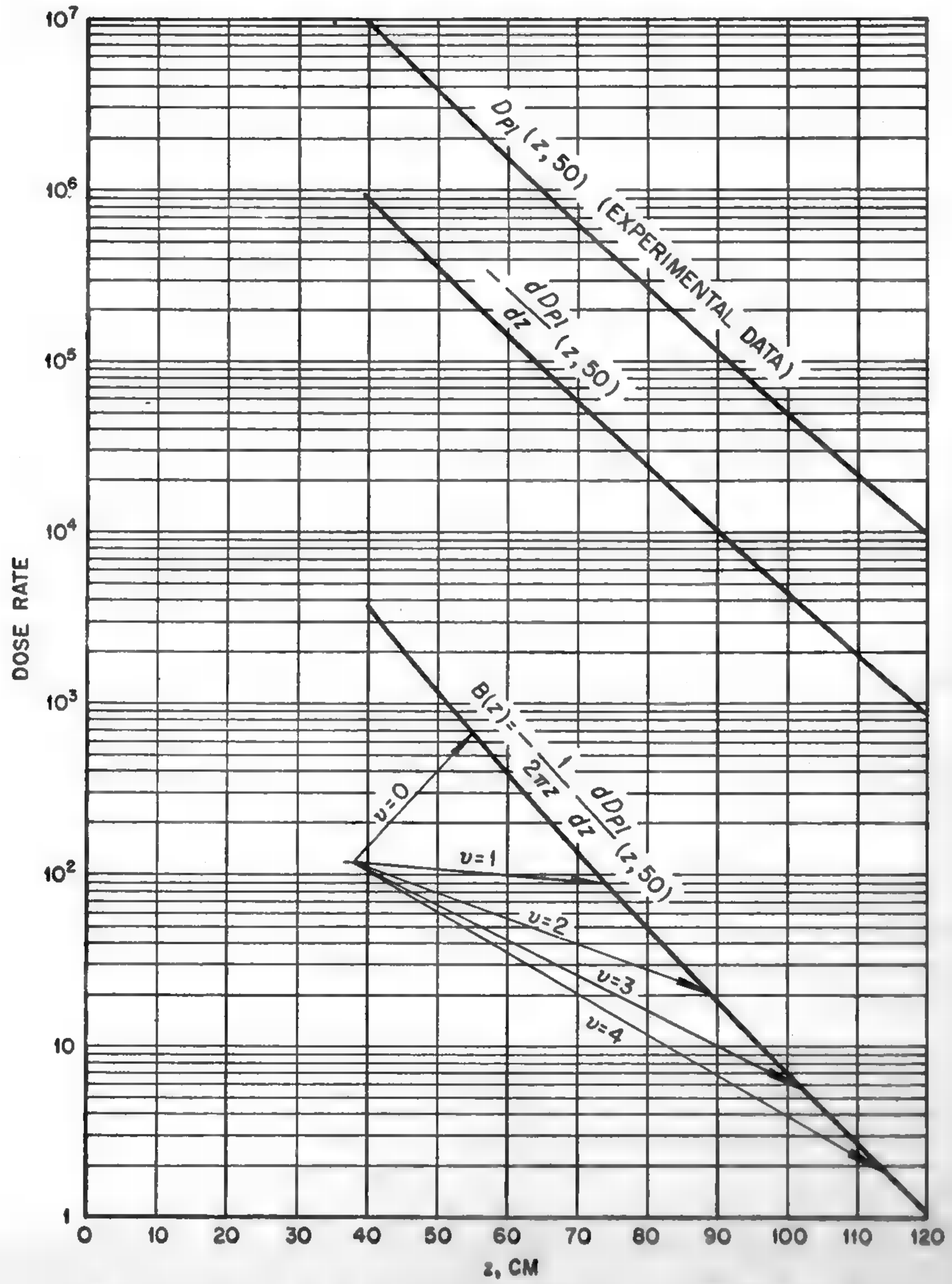


FIG. 10.12. Dose rates from circular disk source (E. P. Blizard)

source, and $D_{pl}(z, a)$ may be taken as equal to $D_{pt}(z)$, from which $G(z)$ is obtained directly by definition (§ 10.52).

10.135. The use of equation (10.134.2) may be illustrated by means of Fig. 10.12. The top curve represents experimentally determined dose rates at distances, z, ranging from 40 to 120 cm along the axis of a circular disk source of radius $a = 50$ cm. The next lower curve gives the negative slope of the $D_{pl}(z, 50)$ curve as a function of z, whereas the third curve represents the corresponding values of $B(z)$ for unit source strength, i.e., for $S = 1$. Taking z as 55 cm, the arrows point to the values of $B(\sqrt{z^2 + \nu a^2})$ for $\nu = 0$, 1, 2, 3, and 4, respectively. It is seen that the convergence is quite rapid, so that it is not necessary to go beyond $\nu = 4$. The sum of the five B terms is about 790, and this gives the point kernel $G(z)$ for $z = 55$ cm.

10.136. An analogous transformation to that given above may be used to relate the observed dose rates for a finite disk source to those to be expected from an infinite plane source. By combining equations (10.132.1), (10.134.1), and (10.134.2) and integrating, it is found that

$$D_{pl}(z, \infty) = D_{pl}(z, a) + D_{pl}(\sqrt{z^2 + a^2}, a) + D_{pl}(\sqrt{z^2 + 2a^2}, a) + \cdots$$
$$= \sum_{\nu=0}^{\infty} D_{pl}(\sqrt{z^2 + \nu a^2}, a). \qquad (10.136.1)$$

Consequently, if the dose rates recorded from a disk source of radius a at distances z, $\sqrt{z^2 + a^2}$, $\sqrt{z^2 + 2a^2}$, $\sqrt{z^2 + 3a^2}$, etc., from the source are added, the sum will give the dose rate at z from an infinite plane source having the same strength per unit area.* The series converges rapidly when a is large in comparison with the relaxation length of the radiation.

10.137. In some circumstances it is not possible to obtain a sufficient number of reliable measurements to perform the summation required by equations (10.134.2) and (10.136.1). An approximate relationship, which is derived from equation (10.136.1), can then be used, provided the series converges rapidly and the dose rate in the region of interest can be expressed in the form of a simple exponential equation in terms of an apparent relaxation length λ. It is then found that

$$1 + \alpha > \frac{D_{pl}(z, \infty)}{D_{pl}(z, a)} \geq \tfrac{1}{2} + \alpha, \qquad (10.137.1)$$

where

$$\alpha \equiv \frac{2\lambda^2}{a^2}\left(\frac{z}{\lambda} + 1\right). \qquad (10.137.2)$$

This is the relationship given, without proof, in § 10.78. In general, the lower limit, i.e., $\frac{1}{2} + \alpha$, is a very good approximation to the ratio of the dose rates from infinite and finite plane (disk) sources.

* This procedure is known as the *Hurwitz transformation.*

10.138. The use of the transformation of equation (10.137.1) requires a knowledge of the apparent relaxation length of the radiation in the absorbing medium. This can be obtained from the data in Fig. 10.12. From the general definition of relaxation length [equation (10.65.1)],

$$\frac{1}{D_{p1}(z, a)} \cdot \frac{d}{dz} D_{p1}(z, a) = -\frac{1}{\lambda},$$

and so λ at any point z is readily obtained upon dividing the value of $D_{p1}(z, a)$, from the top curve, by the corresponding $-dD_{p1}(z, a)/dz$, from the second curve. Thus it is found that, for $z = 55$ cm, λ is about 11 cm. Since a, the radius of the disk source to which Fig. 10.12 applies, is 50 cm, the value of α, defined by equation (10.137.2), is 0.58. Consequently from equation (10.137.1)

$$\frac{D_{p1}(55, \infty)}{D_{p1}(55, 50)} \approx 1.08.$$

From Fig. 10.12, $D_{p1}(55, 50)$ is about 2.2×10^6, so that $D_{p1}(55, \infty)$ is approximately 2.4×10^6. This result is somewhat low, as may be seen by applying equation (10.136.1) to Fig. 10.12. Upon adding the values of $D_{p1}(z, 50)$, from the top curve, corresponding to the points for $\nu = 0$, 1, 2, etc., it is found that $D_{p1}(55, \infty)$ is somewhat greater than 2.7×10^6. For many rough calculations the difference between 2.4×10^6 and 2.7×10^6 is not very significant.

Generalized Curved Surface Source

10.139. In the following section there will be derived an expression for the dose rate from a generalized curved surface source. In two special cases, namely, when the surface is that of (1) a sphere or (2) a right circular cylinder, the results obtained reduce to the expressions given earlier.

10.140. Consider an isotropic radiation source, spread uniformly over a surface; let the point O on the surface nearest to the detector be the origin. The xy-plane at the origin is tangent to the surface (Fig. 10.13). For a point, such as P, not far from the origin, let the distance z_1 from the xy-plane be given approximately by

$$z_1 \approx \frac{1}{2}\left(\frac{x^2}{a} + \frac{y^2}{b}\right), \tag{10.140.1}$$

where a and b are the normal curvatures at the surface. Although this approximation does not apply to points at an appreciable distance from O, very little error is introduced by assuming the expression to hold for the whole surface. This is because most of the radiation received at the detector comes from sources near the origin, whereas the contribution of the more distant sources is much less important.

10.141. The dose rate received at a distance z from the origin is represented by

$$D(z) = S \int_{\text{surface}} G(R)\, ds, \tag{10.141.1}$$

where ds is a small element on the surface, S is the uniform source strength per unit area, and $G(R)$ is the point kernel; the integration is carried over the whole of the surface. The value of ds is given by

$$ds = dx\, dy \sqrt{\left(\frac{\partial z_1}{\partial x}\right)^2 + \left(\frac{\partial z_1}{\partial y}\right)^2 + 1}$$
$$\approx dx\, dy \left(\frac{x^2}{2a^2} + \frac{y^2}{2b^2} + 1\right), \tag{10.141.2}$$

using equation (10.140.1) for z_1.

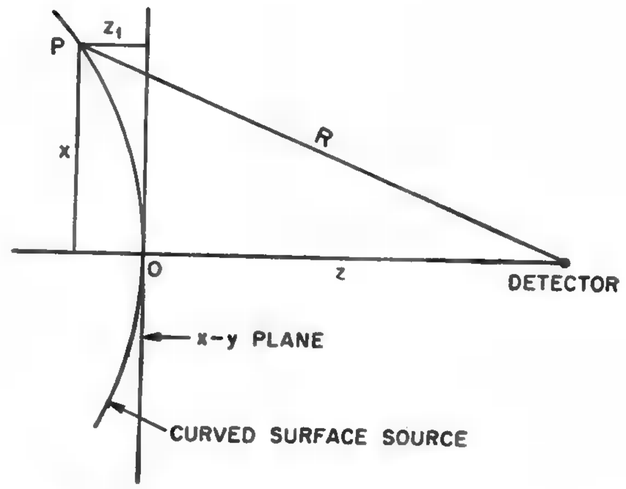


Fig. 10.13. Generalized curved surface source

10.142. It is now postulated that $G(R)$ can be expressed as the product of an unspecified, and presumably exact, kernel $G(z)$ for the distance z, and an exponential term for the additional distance $R - z$; thus

$$G(R) = G(z)e^{-(R-z)/\lambda}, \tag{10.142.1}$$

where λ is the apparent relaxation length which makes this expression correct. Actually, λ will vary slowly with distance, but a satisfactory value for the region of interest can be obtained from experimental data. Upon combining equations (10.141.1), (10.141.2), and (10.142.1), the result for an infinite surface is

$$D(z) = SG(z) \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} e^{-(R-z)/\lambda} \left(\frac{x^2}{2a^2} + \frac{y^2}{2b^2} + 1\right) dx\, dy. \tag{10.142.2}$$

In order to perform the integration, $R - z$ must be expressed in terms of x, y, and z. This is done by writing

$$R = \sqrt{(z_1 + z)^2 + x^2 + y^2}$$

and substituting the approximate value $R - z$ for z_1; upon expanding and using

equation (10.140.1) it is found that

$$R - z \approx \frac{x^2}{2z} + \frac{y^2}{2z} + \frac{x^2}{2a} + \frac{y^2}{2b}.$$

Upon inserting this into the exponential term in equation (10.142.2), the integration can be performed analytically. If a and b are large compared with λ, the result reduces to

$$D(z) \approx 2\pi SG(z) \frac{\lambda z \sqrt{ab}}{\sqrt{(a+z)(b+z)}} \quad \text{for} \quad a, b \gg \lambda. \tag{10.142.3}$$

10.143. For the case of a *spherical surface source*, $a = b = r$, the radius of the sphere, and z is equivalent to $r_0 - r$, as may be seen by comparing Fig. 10.3 and Fig. 10.13; consequently equation (10.142.3) now becomes

$$D_{sp}(r_0, r) \approx 2\pi SG(z)\lambda z \frac{r}{r_0} \tag{10.143.1}$$

or, in view of equation (10.132.1),

$$D_{sp}(r_0, r) \approx -\frac{d}{dz} D_{p1}(z, \infty)\lambda \frac{r}{r_0}. \tag{10.143.2}$$

If the relaxation length λ employed above for the spherical surface applies also to the infinite plane source, then, since $d \ln D_{p1}(z, \infty)/dz$ will be equal to $-1/\lambda$, it follows that

$$-\lambda \frac{d}{dz} D_{p1}(z, \infty) = D_{p1}(z, \infty)$$

and, consequently, from equation (10.143.2),

$$D_{sp}(r_0, r) \approx \frac{r}{r_0} D_{p1}(z, \infty)$$

$$\approx \frac{r}{r_0} D_{p1}(r_0 - r, \infty),$$

which is exactly equivalent to equation (10.60.3).

10.144. The transformation for a *cylindrical surface source* can be derived in an analogous manner; this is achieved by setting $a = \infty$, $b = r$, and $z = r_0 - r$ in equation (10.142.3), where r is the radius of an *infinitely long* cylinder and r_0 is the distance from its axis to the detector (cf. Fig. 10.4 and Fig. 10.13). The result is

$$D_{cy}(r_0, r) \approx 2\pi SG(z)\lambda z \sqrt{\frac{r}{r_0}},$$

and, by using the same procedure as with equation (10.143.1), it is seen that

$$D_{cy}(r_0, r) \approx \sqrt{\frac{r}{r_0}} D_{p1}(r_0 - r, \infty),$$

which was given as equation (10.61.1).

Fractional Escape of Radiation from Self-absorbing Source*

10.145. For certain calculations, particularly in connection with the heat generated in reflector and shield, it is desirable to know the fraction of a particular radiation that escapes from the reactor core. This is calculated in the following manner. Consider an infinite slab source of finite thickness H, and let S be the strength in particles per cm³ per sec of a given radiation. Then from the arguments in § 10.82, using Fig. 10.7, it follows that for particles originating in the annulus distant R from the point P on the surface, the component at right angles to the exterior surface is given by

$$\text{Particles from annulus incident upon 1 cm}^2 \text{ of surface per sec} = 2\pi SG(R)R^2 \sin\theta \cos\theta \, d\theta \, dR,$$

where $G(R)$ is the point kernel; the $\cos\theta$ term is introduced because the component at right angles to the surface is required. The total radiation incident on the surface is now obtained by integrating over all values of θ from zero to $\pi/2$ and of R from zero to $H/\cos\theta$; thus, representing $G(R)$ by equation (10.69.1) with the build-up factor equal to unity, it follows that

$$\begin{aligned}\text{Total number of particles incident upon 1 cm}^2 \text{ per sec} &= \frac{S}{2}\int_0^{\pi/2} \sin\theta\cos\theta \, d\theta \int_0^{H/\cos\theta} e^{-R/\lambda} \, dR \\ &= \frac{S\lambda}{2}\int_0^{\pi/2} \sin\theta\cos\theta(1 - e^{H/\lambda\cos\theta}) \, d\theta \\ &= \frac{S\lambda}{2}\left[\frac{1}{2} - F_2\left(\frac{H}{\lambda}\right)\right],\end{aligned}$$

where $F_2(q)$ is the integral function defined by

$$F_2(q) \equiv q^2 E_3(q),$$

and $E_3(q)$ is defined by equation (10.71.2).

10.146. Within the slab source the number of particles incident upon 1 cm² per sec is SH, and so

$$\begin{aligned}\text{Fraction of radiation escaping from sides of slab} &= \frac{\frac{S\lambda}{2}\left[\frac{1}{2} - F_2\left(\frac{H}{\lambda}\right)\right]}{SH} \\ &= \frac{\lambda}{2H}\left[\frac{1}{2} - F_2\left(\frac{H}{\lambda}\right)\right]. \end{aligned} \tag{10.146.1}$$

For a long cylindrical source of radius R, the corresponding value would be

$$\text{Fraction of radiation escaping from surface of cylinder} = \frac{\lambda}{R}\left[\frac{1}{2} - F_2\left(\frac{R}{\lambda}\right)\right]. \tag{10.146.2}$$

Example The core of a Swimming Pool type reactor is equivalent to a cylinder of 21 cm radius. If the relaxation length of 3-Mev gamma radiation in the core is 23 cm, what fraction of this radiation escapes from the exterior?

* This derivation was included at the suggestion of J. A. Lane; the result is used in Chapter XI.

Substituting 21 cm for R and 23 cm for λ in equation (10.146.2) it follows that

$$\text{Fraction of 3-Mev radiation escaping} = \tfrac{23}{21}[\tfrac{1}{2} - F_2(\tfrac{21}{23})]$$
$$= 1.1[0.5 - F_2(0.9)].$$

From tables it is found that $F_2(0.9)$ or $(0.9)^2E_3(0.9)$ is equal to 0.12, so that

$$\text{Fraction of 3-Mev radiation escaping} = 1.1(0.5 - 0.12)$$
$$= 0.42.$$

Symbols Used in Chapter X

A	atomic weight, mass number
a	radius of disk source
a, b	normal curvatures of surface
$B(R)$	build-up factor
$B(z)$	function defined by equation (10.134.1)
C	number of curies
D	radiation dose rate
E_n	exponential integral function defined by equation (10.71.2)
e	base of natural logarithms
$G(R)$	point kernel for distance R
$G(z)$	point kernel for distance z
g	mass of radioactive species in grams
H	thickness of slab source
$I(0)$	radiation intensity before passage through shield
$I(R)$	radiation intensity after passage through thickness R of shield
l	replacement length for gamma rays
n	integer
q	variable
R	nuclear radius
R	radius of cylinder
R	distance from radiation source to detector (or shield thickness)
r	radius of spherical surface source
r	radius of cylindrical surface source
r_0	distance from center of sphere to detector
r_0	distance from axis of cylinder to detector
S	source strength
S_a	equivalent surface source strength
S_v	volume source strength
t	thickness of gamma-ray shield
$t_{\frac{1}{2}}$	half life of radioactive species
x, y, z	coordinates
z	thickness of absorber (shield)
α	function defined by equation (10.78.2)
Γ	gamma-ray dose rate

γ	number of gamma-ray photons per decay
θ	angle
λ	relaxation length of radiation (approximately equivalent to $1/\mu$ or $1/\Sigma$)
μ	linear absorption coefficient for gamma rays
ν	zero or integer
ρ	radius of annulus
ρ	density
σ_a	microscopic absorption cross section
σ_r	microscopic effective removal cross section for fast neutrons
Σ	macroscopic absorption cross section
Σ_r	macroscopic effective removal cross section for fast neutrons
τ	Fermi age of thermal neutrons
ϕ	flux

PROBLEMS

1. After traversing a shield 6 ft thick, in which the relaxation length is 10 cm, the radiation dose rate received from a spherical surface source, radius 3 ft, was equivalent to 1 mrem/hr. For the same shield, what would be the dose rate from a plane circular source, radius 3 ft, of the same strength per unit area, located at the center of the sphere?

2. A reactor behaves like a plane isotropic source emitting 10^{10} gamma-ray photons/(cm^2)(sec). Estimate the flux at a detector located behind a shield of 7 ft of barytes concrete (density 3.5 g/cm^3), assuming a linear build-up factor.

3. A shield is to be built around a cylindrical tank, 4 ft high and 2 ft radius, to contain a radioactive liquid. The tank may be erected so that it either stands vertically on its base or lies horizontally on its side. Compare the shield thicknesses required to give the same dose rate in the two cases. What is the physical basis of the difference?

4. A horizontal cylindrical tank, 6 ft long and 3 ft diameter, is to contain an aqueous solution of 100 curies of iodine-131; this isotope emits a 0.36-Mev gamma photon in 85 per cent of decays and a 0.64-Mev photon in the other 15 per cent. What is the thickness of an iron shield that would be required to reduce the gamma-ray dose rate from the end of the tank to the maximum permissible value for a 40-hr week exposure?

5. The dimensions of the core of a Swimming Pool type reactor are 1 ft $\times$ 1 ft $\times$ 2 ft high; it may be regarded as consisting of water and aluminum in the volume ratio of 3.3, together with 3.5 kg of uranium. Calculate the minimum depth of water which would be required in the pool to act as a shield over the top of the reactor for operation at 100 kw.

6. In the preceding problem, what thickness of water would be required at the sides of the reactor in addition to a 2-ft thick wall of ordinary concrete?

7. A Water Boiler reactor contains 0.9 kg of uranium, as uranyl nitrate, dissolved in 13 liters of water in a thin-walled steel sphere of 1 ft radius. The sphere is surrounded by a graphite reflector having a thickness of 4 ft 6 in., and a shield of ordinary concrete 5 ft thick. Determine the dose rate in mrem/hr received through the shield when the reactor operates at a power of 45 kw.

8. Estimate the effective removal cross section of magnesium for fast neutrons, and compare the masses and thicknesses of magnesium and iron required to attenuate fast neutrons to a specified extent in a water-containing shield.

9. Calculate the fast-neutron dose rate to be expected through a 3-ft thick shield, of the same material as that to which Fig. 10.11 applies for the LTSF, from a spherical reactor of 100 cm radius having an equivalent isotropic surface source strength of 5 watts/cm².

10. From Fig. 10.12 determine the values of the point kernel $G(z)$ for $z = 45$ and 65 cm. By combining these values with the result in the text for $z = 55$ cm, derive an analytical expression for $G(z)$ as a function of z.

Chapter XI *

THERMAL ASPECTS OF NUCLEAR REACTOR SYSTEMS

THERMAL PROBLEMS IN REACTOR DESIGN

Introduction

11.1. It has been seen earlier (§ 1.28) that there is no theoretical upper limit to the rate of energy release, i.e., of power production, due to fission. In practice, however, the maximum power level of a reactor is frequently determined by the rate at which heat can be removed. Thus, in nuclear reactors operating at high neutron fluxes, such as those intended for central-station power, for ship or aircraft propulsion, or for plutonium production, the design of the core may depend just as much on the heat-removal aspects as on nuclear considerations. The transfer of heat from fuel elements to the coolant is facilitated by increasing both the contact area and the coolant-channel volume. However, such increases generally result in a decrease of the multiplication factor of the system, so that additional fissionable material is required to make the reactor critical. The heat-transfer and nuclear requirements may consequently be in conflict, and the actual design may represent a compromise between the opposing factors.

11.2. Most existing reactor-design concepts represent particular solutions of the heat-transfer problem in which the choice of coolant, the arrangement of fuel and coolant, and the method of heat removal are the primary considerations. Although each type of reactor has its own specific thermal problems, the solution of these problems can be approached in all cases by the application of conventional engineering principles of heat transfer, hydrodynamics, and thermal stress. The equations needed to analyze the thermal behavior of a reactor are, therefore, those concerned with temperature distribution and thermal stress in solid components and with the flow of reactor coolants or liquid fuels. It is the purpose of this chapter to show how these equations are derived and to indicate how they are applied to the design of the various components of the heat-removal system of a reactor.

* Material contributed by S. Glasstone, J. A. Lane, N. F. Lansing, R. N. Lyon, H. F. Poppendiek, and P. C. Zmola.
Reviewed by J. A. Lane, N. F. Lansing, and H. F. Poppendiek.

11.3. Before considering the individual problems, however, mention should be made of several unusual thermal aspects of nuclear reactor design. The first of these involves the temperature of the system and its relation to the rate of heat generation and removal. Since heat is being continuously produced by fission and other nuclear processes, a path must be provided for its transmission to a sink to prevent a steady temperature increase. Unlike a conventional power plant, where the temperature is limited to that resulting from the combustion of coal, oil, or gas, the temperature in a nuclear reactor can rise to a value which depends either upon the rate of heat removal or upon the physical integrity of the materials in the reactor. An example of the latter possibility would be an uncooled fuel element, in which the heat from a sustained fission reaction raises the temperature to the point where the element melts or vaporizes and destroys itself and its container. In practical cases, however, the temperature in the fuel elements and other parts of the reactor structure are determined by the rate of heat generation, the resistance to heat flow from the elements to the coolant, and the temperature of the coolant itself.

11.4. For a reactor of given design, the maximum operating power is limited by some temperature in the system. This limit may be set by the phase stability of the fuel elements, the coolant, or the moderator, by allowable thermal stresses in some parts of the system, by the influence of temperature on corrosion, or by other thermal effects. The maximum permissible temperature must be definitely established to make sure that the cooling system is adequate under all anticipated operating conditions. This is done by, first, estimating the magnitude and distribution of the heat sources in the reactor system and, second, determining the temperature differences along the various paths of heat flow in the system. These two interrelated areas of investigation form the basis for both the concept of a new reactor system and the thermal analysis of a specific design.

Generation and Disposal of Heat in Reactor Systems

11.5. The determination of the manner in which the heat sources are distributed in the reactor core and structure is complicated by the fact that the heat is generated in a number of different ways. The major portion arises from the kinetic energy of the fission fragments which usually manifests itself as heat produced entirely within the fuel elements, but heat is also produced from the slowing down of neutrons and beta particles, and the absorption of various gamma radiations. Since the neutrons and gamma rays are not uniformly distributed throughout the reactor core and structure, the associated heat-source distribution will also be nonuniform. An accurate knowledge of neutron and gamma-ray fluxes is therefore necessary for a complete solution of the reactor cooling problem. These fluxes are difficult to estimate with sufficient accuracy for thermal calculations, and so, in the final analysis, the reactor designer must frequently rely upon data obtained from experiments or from operating reactors.

11.6. Once the heat-source distribution is known, the determination of the temperature distribution in the reactor and of the temperatures of the various coolant circuits leading to the heat sink can be made in a straightforward manner. Here the main objective is to follow the heat flow from the fuel element to the coolant and from the coolant to a sink. Although a heat sink must be provided in all cases, the heat-flow path will vary with the nature of the reactor system. In some cases the heat is discharged directly from the primary coolant to a sink, e.g., a river or the atmosphere. In other instances, a secondary or even a tertiary coolant may be used to transfer heat from the primary coolant to the sink. Further reference to this aspect of the design of the thermal system of a reactor will be made shortly.

11.7. The temperature at any point in a reactor will obviously be greater than that of the sink by an amount equal to the sum of all the temperature drops along the heat-flow path. The temperature drops will usually be proportional to the amount of heat flowing and, hence, to the reactor power. Thus, the steady-state temperature in any component is generally determined by the sink temperature, the reactor power level, the total power generated in the component, and the effective thermal resistance between the component and the sink.

Special Thermal Problems

11.8. In addition to the manner in which heat is generated and disposed of in a reactor, there are other special problems, unique to reactor design, which must be considered. There is, in particular, the matter of selecting reactor materials and coolants, which was discussed in Chapter VIII. In a conventional heat engine or power plant, materials of construction are chosen on the basis of mechanical performance, but in a reactor system the choice is dictated, to a considerable extent, by nuclear properties. This often results in the selection of unconventional, high-cost materials with their attendant problems. Such materials do not always have the most desirable thermal, physical, or mechanical properties, and frequently these properties are not well known. Beryllium metal, for example, is an excellent material for use as moderator and reflector, but it is relatively brittle (§ 8.161). Consequently great care must be taken when using beryllium to avoid high temperature differences which would give rise to excessive thermal stresses and thus cause cracking.

11.9. Another factor which adds to the problem of heat removal from a reactor is that the volumetric heat-release rates may be higher than for any other thermal systems designed for continuous operation. There are two reasons for this: (1) the necessity for keeping the size of the reactor small, e.g., in fast reactors and in all types of mobile reactors, and (2) the desire to increase the power output per unit mass of fuel (or other expensive reactor materials) in order to reduce ultimate power costs (see § 12.227). In Table 11.1 some typical volumetric heat-release rates (or power densities) in reactors are com-

pared with the values for other thermal sources. The data for reactors are based on the total core volume occupied by fuel.

TABLE 11.1. COMPARISON OF VOLUMETRIC HEAT-RELEASE RATES

Thermal Sources	Watts/cm³ (or kw/liter)	$10^{-4} \times$ Btu/(hr)(ft³)
Brookhaven reactor (28 Mw)	0.06	0.58
Steam boiler: natural convection	0.5	4.8
Steam boiler: forced convection	10	97
Homogeneous Reactor Experiment (1 Mw)	20	190
Aircraft gas-turbine combustion chamber	44	400
Experimental Breeder Reactor (~1 Mw)	240	2,300
Materials Testing Reactor (30 Mw)	300	2,900
Typical power reactor (estimated)	600	5,800
V-2 rocket combustion chamber	20,000	193,000

11.10. The combination of high operating temperatures (for maximum thermal output of the reactor and maximum thermodynamic efficiency for ultimate conversion into power) and large volumetric heat sources makes special demands upon the design of the reactor heat-removal system. Not only do the large source densities cause stresses due to temperature differences within core components to increase rapidly as their thicknesses increase, but the nuclear radiations at high power densities, e.g., high neutron fluxes, may have adverse effects on the thermal conductivity and other properties of reactor materials (see Chapter VIII). In general, the major advances in reactor design have come from material and fabrication developments, but it is only by optimizing the reactor systems from the heat-transfer standpoint that their full advantage can be realized. A large part of the improvement in reactors can therefore be attributed to good heat-transfer design.

DESIGN OF COOLING SYSTEM

Coolant Circuits

11.11. The simplest solution of the reactor heat-removal problem is to employ a direct-cycle cooling system in which the coolant is passed once through the reactor and then to a sink. In the Hanford and Chalk River (NRX) reactors, for example, river water is the coolant and after passage through the reactor it returns to the river, which acts as the sink. The Oak Ridge (X-10) and Brookhaven natural-uranium reactors, and the British (Windscale) production reactors use air as coolant and the atmosphere as the sink. These systems, although simple in principle, are not without problems. River water must be purified before it is acceptable as a reactor coolant (§ 8.236), and provision must be made for preventing the discharge of radioactive air or water to the surroundings as described in Chapter IX.

11.12. The use of a secondary coolant system in conjunction with a closed primary-coolant circuit has the advantage of better control over the escape of radioactivity. Such systems are considerably more flexible than those using a direct cycle, particularly with respect to location of the reactor and choice of primary coolant. Heavy water, for example, is a desirable coolant from the standpoint of its nuclear properties, but its cost necessitates a closed primary circuit, so that a secondary coolant, e.g., ordinary water, is required to transfer the heat to the sink. Cooling systems of this type, i.e., heavy water as primary coolant in a closed circuit and ordinary water in an open circuit, are used in the CP-3′ and CP-5 reactors at the Argonne National Laboratory, and in other reactors of similar design (see Chapter XIII).

11.13. Even when the coolant is ordinary water, but highly purified, as in the Materials Testing Reactor (MTR), it may be desirable for reasons of economy to use a secondary cooling system. The secondary coolant may be either air or water and the heat sink will be either the atmosphere or a river.

11.14. In the systems considered above, little or no use is made of the heat generated by fission. In reactors designed for power production, however, three successive separate coolant circuits may be necessary. In most reactor power system designs the primary coolant is used to boil water in a secondary circuit and thus produce steam. The steam then drives a turbine, the water from which is condensed by means of a third coolant circuit which discharges heat to the sink. A boiling-water power reactor, however, may not require the three coolant circuits, since the steam from the primary circuit may be used directly in the turbine, so that the secondary system is eliminated.

11.15. In a reactor using sodium (or NaK) as coolant it may be desirable to interpose a liquid, such as mercury, molten lead, or an organic compound, between the sodium and boiling-water (steam) loops to avoid the possibility of chemical reaction between these two materials. The use of an intermediate system of this type can also serve the purpose of restricting the volume that has to be shielded because of the gamma radiations from sodium-24 produced by neutron capture in the reactor. A sodium-cooled power reactor may thus have four different coolant circuits, e.g., sodium, mercury, boiling water, and condenser water.

11.16. The use of a circulating liquid fuel, as in the Homogeneous Reactor Experiment (HRE), simplifies, in some respects, the heat-transfer problem because the heat produced by fission is removed outside, rather than inside, the reactor (see § 12.132). However, circulating liquid-fuel systems have their own particular problems, such as the design of the primary heat exchanger in which heat is transferred from the liquid fuel to water for the production of steam in the secondary circuit. Because of the intense radioactivity of the circulating fuel, due to the presence of fission products, the heat exchanger must be constructed from corrosion-resistant materials, and be absolutely reliable and leak-proof. It should also have a small volume in order to minimize the hold-up

(and inventory) of fuel solution. If the liquid fuel is under pressure, e.g., about 1000 psi in the HRE, the construction specifications of the heat exchanger become even more stringent.

Heat-Source Distribution

11.17. In a reactor using solid fuel, the design of the fuel elements and of the spacing of coolant passages through the various solid components is influenced by a determination of the temperature distribution within these components. The first step in such a determination is to establish the maximum volumetric heat-release rate in each of the components being considered. The procedure for doing this will be discussed below. Although the heat-source distribution depends upon the particular reactor design, there are certain generalizations which can be used for preliminary calculations, at least.

11.18. It may be assumed that, in a thermal reactor, 90 per cent of the total heat is liberated in the fuel elements, and that this heat source will be distributed in a manner proportional to the neutron flux. About half of the remaining heat, i.e., 5 per cent of the total, will be released in the moderator, and the remaining 5 per cent in the reflector and shields. The moderator heat-source distribution will be somewhere between a uniform distribution and one proportional to the thermal-neutron flux. In many reactor designs, each cooling channel takes up heat from both the fuel element and the surrounding moderator, and in these cases, the two heat sources are combined.

11.19. The determination of the heat-source distribution in the reflector and shields is somewhat complicated, because it involves an estimate of the neutron leakage from the core and of the gamma-radiation flux as a function of the energy spectrum. Since gamma rays arise from a number of sources, e.g., fission, fission-product decay, and radiative capture (cf. § 10.9, *et seq.*), the details of each originating nuclear process must be known and the subsequent behavior of the radiations calculated. Although only 5 per cent of the total heat is released in the reflector and shields, including the pressure vessel if one is used, the cooling of these components may be a serious problem in a reactor operating at high power.

Temperature Distribution

11.20. The second step in analyzing or designing the cooling system of a reactor is to calculate the temperature distribution in the solid components from their known heat-source distributions, thermal conductivities, and configurations, using appropriate heat-flow equations. Since fuel elements are usually in the form of cylinders, hollow rods, or parallel plates, the heat flow and temperature distribution in these cases can be determined in a relatively simple manner, as will be seen below.

11.21. Solid moderator and reflector components, on the other hand, do not always have simple configurations; further, the spacing of cooling channels in

the shields may not be uniform, because the heat liberated falls off with increasing distance from the core. Such cases can often be treated by simplifying the geometry to a form, e.g., a cylinder, which permits the use of an established heat-flow equation. Alternatively, a point-to-point analysis can be made using a relaxation technique.*

11.22. The design of a reactor cooling system next requires the determination of the flow characteristics and heat-transfer coefficients of the coolant. This fixes the temperature drop between the surface of the solid being cooled and that of the bulk of the coolant, as well as the temperature rise in the coolant stream. The increase in temperature of the coolant flowing through the hottest channel is of special significance, since this establishes the maximum temperature of the system.

11.23. Finally, it is necessary to consider the transfer of heat, in stages, from the primary coolant to the sink. This may involve heat exchange from the primary coolant to a steam system, or from the primary coolant to a secondary coolant and then to a steam system, and ultimately to a heat sink. This aspect of the subject is treated along more or less conventional lines.

Auxiliary Thermal Problems

11.24. In addition to the matters dealt with above, there are a number of auxiliary problems, related to the thermal characteristics of a reactor, that have a bearing on the design of the cooling system. One of the most important of these is the problem of stress in solid components, due to temperature differences. For example, problems of stress may arise in the design of thermal and biological shields, because of the considerable difference of temperature between the two faces. Further, in pressurized-water reactors, thermal stresses in the thick-walled pressure vessel become very significant.

11.25. The coolant itself often presents special problems, which have been examined in Chapter VIII, and which may be reviewed briefly here. Mention may be made of purification, of induced radioactivity resulting from neutron capture, of decomposition by radiation, and of corrosion (or erosion), including mass transfer. These are highly important aspects of the design of the thermal system of a reactor, but they represent problems in materials and equipment rather than of heat transfer.

11.26. In summarizing the foregoing discussion it can be seen that the design or analysis of the cooling system of a reactor involves the following topics:

(1) Heat sources and their distribution.
(2) Heat conduction through solid components and to the primary coolant (or heat convection within liquid fuels).
(3) Heat transport along reactor channels due to flow of fluid.

* H. W. Emmons, *Trans. Am. Soc. Mech. Eng.*, **65**, 909 (1943); P. B. Richards, *Chem. Eng. Prog. Symposium Series*, No. 11, **50**, 127 (1954).

(4) Heat exchange from primary to secondary coolant and to steam-generating system (or heat sink).
(5) Thermal stresses in solid components.

These are the main subjects which will be considered in the succeeding sections of this chapter.

HEAT SOURCES IN REACTOR SYSTEMS

Spatial Distribution of Fission Energy

11.27. As seen in Chapter I, the energy released during the fission process appears in various forms, but mainly as the kinetic energy of the fission fragments, the fission neutrons, and the beta particles resulting from the radioactive decay of the fission products. The fission fragments are usually stopped within the fuel elements themselves; the small fraction that escapes into the cladding penetrates only about 0.01 mm (0.004 in.). The beta particles of high energy may travel up to 4 mm (0.16 in.) in a cladding material such as aluminum, and so a large fraction of these particles may escape from the fuel element into the moderator or coolant, but they will not get out of the reactor core. The fission neutrons lose most of their energy in the first few collisions with moderator atoms, and they travel distances ranging from some centimeters to a few feet. It is seen, therefore, that most of the heat from the three sources under consideration, comprising about 90 per cent or more of the total energy generated, will be released within the reactor core.*

11.28. The remaining 10 per cent or less of the energy released in fission appears as gamma radiations from the several sources mentioned in § 11.19. The manner in which these are distributed throughout the reactor core, reflector, and shields depends upon the materials present and the configurations of the various components. The distribution will consequently depend upon the reactor type. A complete analysis of the heat generation due to gamma radiation is complicated; it requires a knowledge of the energy spectrum and spatial distribution of each of the gamma-ray sources, as well as the absorption coefficients of the materials in the system for radiations of different energies.

11.29. In addition to the spatial distribution of heat sources, the time dependence must also be considered, since an appreciable proportion of the energy of fission is released over a period of time. For example, the fission fragments may lose essentially all of their kinetic energy in a few microseconds, whereas the energy accompanying the radioactive decay of the fission products is released over many years. Actually, the approach to equilibrium thermal conditions after the start-up of a reactor is very rapid and any departure from this equilibrium can usually be neglected.

* According to the statement in § 11.18, the heat released in the core is about 95 per cent (90 per cent in the fuel elements and 5 per cent in the moderator) of the total. The additional 5 per cent or so arises from gamma-ray absorption in the core.

11.30. However, provision must be made for cooling the fuel elements and other reactor components for some time after shut-down of the reactor because of the heat generated in the decay of the fission products (cf. § 2.180). The source distribution will then be different from that when the reactor is in operation. One hour after shut-down, for example, the heat generation rate in the fuel elements will be about 1 per cent of the operating value, whereas in the reflector and shield it will be approximately 10 per cent of the rate during operation. This difference arises because a large fraction of the heat released after shut-down is due to absorption of gamma rays from fission product decay. Although this is a small proportion of the heat generated in the core during operation, it makes a considerable contribution to the heat liberated in other parts of the reactor system.

Total Energy Release in Fission

11.31. It was seen in Chapter I that the total energy released in fission, and which ultimately appears as heat, is made up of contributions from a number of sources. In general, the total energy, exclusive of the neutrino energy which is lost to the reactor system, may be expressed by

$$E = 191 + E_c \text{ Mev},$$

where E_c is the energy liberated as a result of various parasitic neutron capture processes, e.g., nonfission capture in uranium-235 and uranium-238, and capture in moderator, coolant, structure, etc.; this includes the energy of the capture gamma radiations and the decay energies, i.e., the energies of the alpha and beta particles and gamma rays, of any radioactive species that are formed by parasitic neutron capture.* Since the value of E_c will obviously depend upon the nature of the materials present in the reactor core, it is evident that the total amount of heat produced by fission will vary, to some extent, from one type of reactor to another.

Example Compare the total energy release per fission in a natural uranium-graphite reactor of the X-10 type (§ 13.9) with that in an enriched uranium-water reactor of the Swimming Pool type (§ 13.70), using the following data:

Natural-Uranium Reactor	*U-238*	*U-235*	*Al (Cladding)*	*Graphite*
Neutrons captured per fission†	0.695	0.185	0.005	0.254
Total energy released per neutron capture (Mev)	6.8	6.8	12.6	5.6
Swimming Pool Reactor	*U-238*	*U-235*	*Al (Cladding)*	*Water*
Neutrons captured per fission†	~0	0.185	0.090	0.274
Total energy released per neutron capture (Mev)	~0	6.8	12.6	2.23

* Although E_c should include the decay energies of all radioactive species formed as a result of nonfission neutron reactions, only those of relatively short half life contribute to the energy release in the state of pseudo-equilibrium attained by the reactor soon after start-up.

† Calculated from the amounts of the various species present in the reactor and their respective cross sections.

In the natural-uranium reactor the energy, E_c, liberated as a result of nonfission capture is given by

$$E_c = (6.8 \times 0.695) + (6.8 \times 0.185) + (12.6 \times 0.005) + (5.6 \times 0.254)$$
$$= 7.5 \text{ Mev.}$$

The total energy release per fission is thus $191 + 7.5 = 198.5$ Mev.

For the Swimming Pool reactor type,

$$E_c = (6.8 \times 0.185) + (12.6 \times 0.09) + (2.23 \times 0.274)$$
$$= 2.9 \text{ Mev.}$$

The capture by the small proportion of uranium-238 in the highly enriched fuel makes a negligible contribution. The total energy release is now $191 + 2.9 = 193.9$ Mev per fission. (Direct experimental measurement of the energy release in this reactor gave a value of 193 ± 5 Mev per fission.) *

Heat Generation in Fuel Elements

11.32. The heat generated in the fuel elements containing fissionable material is mainly the sum of that due to the kinetic energy of the fission fragments and the heat resulting from the deceleration of a large fraction of the beta particles and the absorption of a somewhat smaller fraction of the gamma radiation. Although there is a minor dependence upon the design of the fuel elements, it is found, as stated in § 11.18, that up to about 90 per cent of the total reactor heat is released within the fuel.

11.33. The rate of heat liberation, i.e., about 175 to 180 Mev per fission, per unit volume (or power density) will be closely proportional to the rate of fission, i.e., to $\Sigma_f\phi$, per unit volume per second, where Σ_f is the macroscopic fission cross section and ϕ is the neutron flux (§ 2.121). Upon replacing Σ_f by $N\sigma_f$, where N is the number of fissionable nuclei per unit volume of fuel, it is seen that the heat-source distribution in the fuel elements will depend upon N and ϕ, assuming the fission cross section, σ_f, to remain constant. As a general rule, the value of N, which is determined by the distribution of fissionable nuclei, is uniform in a particular reactor. It follows, therefore, that to a good approximation, the thermal source function, expressed as the power density, will have the same spatial dependence as the neutron flux.

11.34. In a heterogeneous reactor in which the lattice consists of widely spaced cylindrical elements of moderately large diameter or of bundles of rods of smaller diameter, the flux distribution within the fuel elements will deviate appreciably from that in the surrounding moderator. Normally, however, the radial flux variation within the elements themselves has little effect on the temperature distribution and so it can be ignored. The main concern is consequently with the over-all flux distribution in the reactor core, and this can be calculated by the methods given in Chapter III.

* J. L. Meem, *Nucleonics*, **12**, No. 5, 62 (1951).

Over-all Average and Maximum Power Densities

11.35. In the practical case, the ratio of the maximum volumetric heat-generation rate (or power density) in the core to the average heat-generation rate is important for heat-transfer calculations. An example of the evaluation of this ratio was given in § 3.217, with the object of showing the effect of a reflector on an infinite slab reactor. The more useful case of a cylindrical reactor will be considered here. The basic assumption of the following calculation is that the fuel is distributed uniformly, but not necessarily homogeneously, throughout the reactor volume.

11.36. From the expression for the neutron flux distribution given in Table 3.7, it can be readily shown that for a right circular cylindrical reactor

$$\frac{\phi}{\phi_0} = J_0\left(2.405\,\frac{r}{R'}\right)\cos\frac{\pi z}{H'}, \tag{11.36.1}$$

where ϕ_0 is the flux at the center of the reactor; J_0 is the zero-order Bessel function of the first kind, r and z are the radial and vertical coordinates, as in Fig. 3.11, and R' and H' are the effective radius and height, respectively, of the fuel-containing portion of the reactor, including an allowance for the reflector. The average neutron flux in the core is given by (cf. § 3.217)

$$\phi_{av} = \frac{1}{\pi R^2 H}\int_0^R\int_{-\frac{1}{2}H}^{\frac{1}{2}H}\phi 2\pi r\,dr\,dz, \tag{11.36.2}$$

where R and H are the actual radius and height of the fuel-containing region. Upon substituting the expression for ϕ from equation (11.36.1) and performing the integration, the result is

$$\frac{\phi_{av}}{\phi_0} = \frac{2RR'J_1\left(2.405\,\frac{R}{R'}\right)}{2.405R^2}\cdot\frac{2H'\sin\left(\frac{\pi}{2}\cdot\frac{H}{H'}\right)}{\pi H}, \tag{11.36.3}$$

where J_1 is the first-order Bessel function.

11.37. Since, as seen above, the power density at any point may be taken as proportional to the neutron flux, it follows that equation (11.36.3) also gives the value of P_{av}/P_0, where P represents the power density. Further, the power density at the center of the reactor, i.e., P_0, is the maximum within the reactor, so that ϕ_{av}/ϕ_0 is essentially equal to P_{av}/P_{max}. For a bare reactor, i.e., one without a reflector, R is approximately equal to R' and H to H'; then, equation (11.36.3), after inversion, becomes

$$\frac{P_{max}}{P_{av}} = \frac{2.405}{2J_1(2.405)}\cdot\frac{\pi}{2} = 3.64.$$

11.38. Calculations similar to the foregoing can be made for reactors of other shapes, and the results for some cores of various geometries are collected in Table 11.2. If, in a cylindrical reactor, the arrangement of fuel elements (or control rods) is such that the radial flux is constant and independent of r, the

TABLE 11.2. RATIO OF MAXIMUM TO AVERAGE POWER DENSITIES IN VARIOUS REACTORS

Core Geometry	P_{max}/P_{av}
Sphere (bare)	3.29
Rectangular parallelepiped (bare)	3.87
Cylinder (bare)	3.64
Cylinder (flat radial flux)	1.57
Swimming Pool type (water reflected)	2.6

value of the ratio P_{max}/P_{av} is just $\frac{1}{2}\pi$, i.e., 1.57. As indicated in § 3.219, the effect of a reflector would be to decrease the ratio for the bare cores.

Average and Maximum Power in Single Fuel Channel

11.39. In reactor cooling problems it is often of interest to estimate the maximum heat-generation rate (or power density) at a point in a given fuel channel, rather than for the whole reactor core. In equation (11.36.1), the first (Bessel function) term gives the radial flux distribution whereas the second (cosine) term represents the longitudinal distribution in a cylindrical reactor. In any specified longitudinal fuel channel, at a fixed radial distance, r, from the reactor center, the neutron flux distribution is

$$\phi_{long} = (\phi_{max})_{long} \cos \frac{\pi z}{H'}, \tag{11.39.1}$$

where $(\phi_{max})_{long}$, the maximum flux at the center of the given channel, is equal to $\phi_0 J_0(2.405r/R')$.

11.40. The average flux in a longitudinal fuel channel in a cylindrical core is given by

$$(\phi_{av})_{long} = (\phi_{max})_{long} \frac{1}{H} \int_{-\frac{1}{2}H}^{\frac{1}{2}H} \cos \frac{\pi z}{H'}\, dz$$

$$= (\phi_{max})_{long} \frac{2H' \sin\left(\frac{\pi}{2} \cdot \frac{H}{H'}\right)}{\pi H}.$$

Hence, assuming the power density to be proportional to the neutron flux, as before, it follows that

$$\left(\frac{P_{max}}{P_{av}}\right)_{long} = \frac{\pi H}{2H' \sin\left(\frac{\pi}{2} \cdot \frac{H}{H'}\right)}. \tag{11.40.1}$$

11.41. The result expressed by equation (11.40.1), or the simplification for the core without end reflectors, is also applicable to fuel channels in a rectangular parallelepiped reactor. For this geometry the flux distribution, in any direction parallel to any of the three principal axes, is represented by a cosine expression, analogous to equation (11.39.1). The only change necessary is to replace H by the actual length of the reactor edge, in the given direction, and H' by the effective length including allowance for the reflector.

More Uniform Power Distribution

11.42. In many reactor designs the power output is limited by the maximum permissible temperature of the fuel elements, i.e., by the value of P_{max}. In some cases it is possible to increase the power output by decreasing P_{max}/P_{av}. In other words, the average power output of the reactor can be increased by a more uniform distribution of the power density. This is sometimes referred to as a "flattening" of the power distribution or neutron flux. From what has been stated above (see also § 3.219), it is obvious that a reflector helps to flatten the neutron flux, but other techniques should also be considered.

11.43. The results obtained in the preceding section are based on the postulate of uniform distribution of the fuel throughout the reactor, but by appropriate nonuniform loading of the reactor the value of P_{max}/P_{av} can be decreased. One way to obtain a relatively flat radial neutron flux is to vary the distribution of fuel and moderator in the reactor.* This procedure has the added advantage of decreasing the mass of fuel required for the attainment of criticality. In cases where the uranium-235 enrichment of the fuel can be varied in different channels, a flat power distribution could be realized by adjusting the concentration of the fissionable species so that it is inversely proportional to the thermal flux at any point. Partial flattening can be achieved in an analogous, but simpler, manner by loading various parts of the reactor with depleted and enriched fuel elements. The elements containing depleted material would be more common near the center of the reactor, whereas more of the enriched fuel elements would be located toward the outside.

11.44. A possible scheme for flattening the flux, and so decreasing the value of P_{max}/P_{av}, is by a suitable distribution of neutron absorbers (or poisons). This could be most easily done by appropriate insertion of control rods, although there is a tendency for the flux to be distorted in the (longitudinal) direction of the rods. A better procedure which has been proposed is to circulate a solution of a poison, e.g., of a cadmium salt, through those regions of the reactor where the neutron flux is usually highest. However, these methods for flattening the flux are uneconomic, since neutrons are used up in a reaction which gives relatively little energy as compared with that released in fission.

11.45. Instead of flattening the flux, as described above, in order to increase the power output of a reactor, the same result, which depends upon a relatively uniform temperature distribution, can be achieved in other ways. These involve variations in the rate of heat removal throughout the reactor, rather than of heat production. The cooling system can be so designed that heat is removed more rapidly from the regions of the reactor, i.e., near the center, where the neutron flux and power production are higher, than from the other regions. One way to do this in reactors with parallel cooling channels is by having entry

* G. Goertzel and W. A. Loeb, *Chem. Eng. Prog. Symposium Series*, No. 12, **50**, 82 (1954); *Nucleonics*, **12**, No. 9, 42 (1954).

orifices of different sizes so that the rate of flow of coolant varies appropriately throughout the reactor. This is sometimes referred to as "orificing" the coolant. An alternative approach, which has been favored by reactor designers, is to use the multipass or multiflow system for the coolant. In the simplest (two-pass) type an arrangement of baffles forces the entering coolant to flow (e.g., upward) only through the central part of the reactor where the power density is highest. The partly heated coolant then flows (e.g., downward) through the outer regions of the reactor where the power density is lower and there is less heat to be removed. A multiflow system involves some increase in the power required to pump the coolant, but this is usually more than offset by its advantages for heat removal.

Heat Generation in Moderator

11.46. The heat produced in the moderator is the result of the slowing down of fission neutrons, the stopping of beta particles from the fission products, and the absorption of gamma rays from various sources. As seen in Chapter I, this constitutes about 5 per cent of the heat generated in the reactor system. Since most of the kinetic energy of the fission neutrons is lost during the first few collisions, the distribution of heat from this source will depend upon the mean free path of the fast neutrons. In reactors where this mean free path is short, e.g., in those having ordinary water as moderator, the heat distribution in the moderator is roughly the same as that of the thermal-neutron flux. If, however, the mean free path is long, the distribution of heat resulting from the slowing down of the fission neutrons will be more nearly uniform throughout the reactor core.

11.47. If the cooling of the moderator is treated as a separate problem from the cooling of the fuel elements, the heat distribution in the moderator must be determined independently. On the other hand, when the cooling of fuel and moderator is combined, the heat-generation distribution in the moderator may be supposed to be similar to the over-all distribution in the fuel.

Heat Generation in Reflector and Shield

11.48. Most of the heat generated in the reflector and shield, amounting to roughly 5 per cent of the total heat produced (§ 11.19), results from the absorption of gamma radiation escaping from the reactor core. The study of the source distribution consequently involves a consideration of the gamma-ray sources and of the absorption of the radiations of various energies in reflector and shields. In many respects determination of the heat-source distribution has features in common with shielding calculations. However, greater precision is usually necessary in the former case. As a general rule, especially for stationary reactors, shields are designed along conservative lines, as seen in Chapter X. Consequently an error as large as 50 per cent or so, for example, in estimating the source strength for shielding calculations might not be at all serious. On the other hand, an underestimate of this extent in evaluating the heat generated in the shield would completely invalidate the design for the cooling system of a power reactor.

11.49. The gamma radiations absorbed in reflector and shield fall into four main categories: (1) prompt gamma rays accompanying fission, (2) fission product decay gamma rays, (3) neutron capture gamma rays, and (4) gamma rays accompanying the decay of radioactive neutron-capture products, if any. These will be considered in turn.

11.50. It was noted in § 2.172 that the investigations of the prompt gamma rays accompanying fission made hitherto do not agree as to the average energy per photon. It appears, however, from empirical studies that for purposes of calculation it may be assumed that five 1-Mev gamma-ray photons are released in the actual fission process.

11.51. During operation of a reactor most of the gamma radiation accompanying the decay of fission products is in the form of photons of relatively high energy, e.g., 2.5 to 3.5 Mev. These arise from radioisotopes with half lives up to a few hours, so that, as far as they are concerned, equilibrium is attained fairly rapidly. A large number of photons of lower energy are also produced from the fission products, but they do not constitute an important heat source. For thermal calculations it is usually sufficient, therefore, to assume that there are two 3-Mev gamma-ray photons per fission, on the average, due to fission-product decay.

11.52. With the single exception of helium-4, every nuclear species present in the reactor system undergoes an (n, γ) reaction, the extent being proportional to the amount of that species present and to its capture cross section. The neutron capture, per fission, in the various components can thus be calculated without difficulty. The total energy of the capture gamma rays is equal to the binding energy of a neutron in the capturing nucleus, but in precise thermal calculations of heat-source distribution in reflector and shields, something about the gamma-ray energy spectrum needs to be known. The data available for a limited number of materials were given in Table 2.6, and these can be utilized for the present purpose, as will be seen below.

11.53. If the product of neutron capture is radioactive, e.g., sodium-24 or aluminum-28, then, in estimating the gamma-ray source, allowance must be made for the gamma radiation accompanying the decay. In this connection radioisotopes of short half life are usually more important than those of long half life.

Heat-Source Distribution in Reflector and Shields

11.54. The first step in evaluating the heat-source distribution in the reflector and shields is to determine the spectrum of the gamma radiation in the reactor core. To do this, the numbers of photons for all energy values should be determined, but in practice the treatment is simplified by restricting the calculations

to a few significant energies. The procedure will be illustrated by means of the following example.

Example Estimate the source of 3-Mev gamma radiations in Mev/(sec)(kw), from a water-moderated reactor of the Swimming Pool type. Photons of this energy arise from fission-product decay and from capture by aluminum and uranium-235.

The energy liberated per fission in this type of reactor is 194 Mev, so that, by the method of § 2.86, there are about 3.2×10^{13} fissions/(sec)(kw). As postulated in § 11.51, the decay of the fission products yields two 3-Mev photons per fission so that this represents a source of $(6)(3.2 \times 10^{13}) = 1.9 \times 10^{14}$ Mev/(sec)(kw) of 3-Mev gamma radiation.

In the example in § 11.31, it was stated that, in the Swimming Pool type of reactor, 0.090 neutron/fission is captured in aluminum; according to Table 2.6, for 3-Mev capture gamma rays, the energy release is 3.8 Mev/capture, so that, in the reactor, the energy is $(0.09)(3.8) = 0.34$ Mev/fission. Consequently, the corresponding source is $(0.34)(3.2 \times 10^{13}) = 0.11 \times 10^{14}$ Mev/(sec)(kw).

Similarly, for uranium-235, there is 0.185 neutron captured per fission, and for 3-Mev gamma rays there are 6.0 Mev/capture. Therefore, this constitutes a source of $(0.185)(6.0)(3.2 \times 10^{13}) = 0.36 \times 10^{14}$ Mev/(sec)(kw).

The total source of 3-Mev gamma-ray photons is thus $(1.9 + 0.11 + 0.36) \times 10^{14} = 2.4 \times 10^{14}$ Mev/(sec)(kw).

11.55. The gamma-ray source spectrum for a reactor of the Swimming Pool type is summarized in Table 11.3. The total Mev/(sec)(kw) for each photon

TABLE 11.3. GAMMA-RAY SOURCE SPECTRA FOR SWIMMING POOL REACTOR CORE

Gamma-ray Source	*Photon Energy* (Mev)	Mev/(sec)(kw)
Fission (prompt)	1.0	1.6×10^{14}
Fission-fragment decay	3.0	1.9×10^{14}
Aluminum capture	3.0	1.1×10^{13}
Aluminum capture	6.0	4.0×10^{12}
Aluminum capture	7.7	7.7×10^{12}
Aluminum-28 decay	1.8	5.2×10^{12}
Water capture	2.2	1.9×10^{13}
Uranium-235 capture	3.0	3.6×10^{13}
Uranium-235 capture	1.0	4.4×10^{12}

energy is obtained by summation of the various contributions, as shown in the example given above.

11.56. The next stage in the calculation is to determine the fraction of the gamma radiations of each energy that escapes from the reactor core. For this purpose use is made of the results of § 10.146, together with the known dimensions of the core and the linear absorption coefficient of the particular radiation in the reactor core. For example, in a Swimming Pool reactor, loaded so that the core is equivalent to a cylinder of 21-cm radius, the fraction of 3-Mev radiation that escapes is calculated as 0.42, so that the rate of escape is $(0.42)(2.4 \times 10^{14}) = 1.0 \times 10^{14}$ Mev/(sec)(kw). This is equivalent to about

55 Btu/(hr)(kw) of 3-Mev gamma radiation. Values for the radiations of other energies can be determined in an analogous manner. These represent the source terms for the subsequent absorption calculations.

11.57. Finally, from the known absorption coefficients of the reflector and shield materials for various gamma-ray energies, it is now possible to calculate the absorption, and hence the heat liberated, at different distances from the reactor core. The values for the several gamma-ray energies are then added to give the total heat-release rate. Some of the results obtained in this manner, per megawatt of power output, are depicted in Fig. 11.1 for a Swimming Pool type reactor.

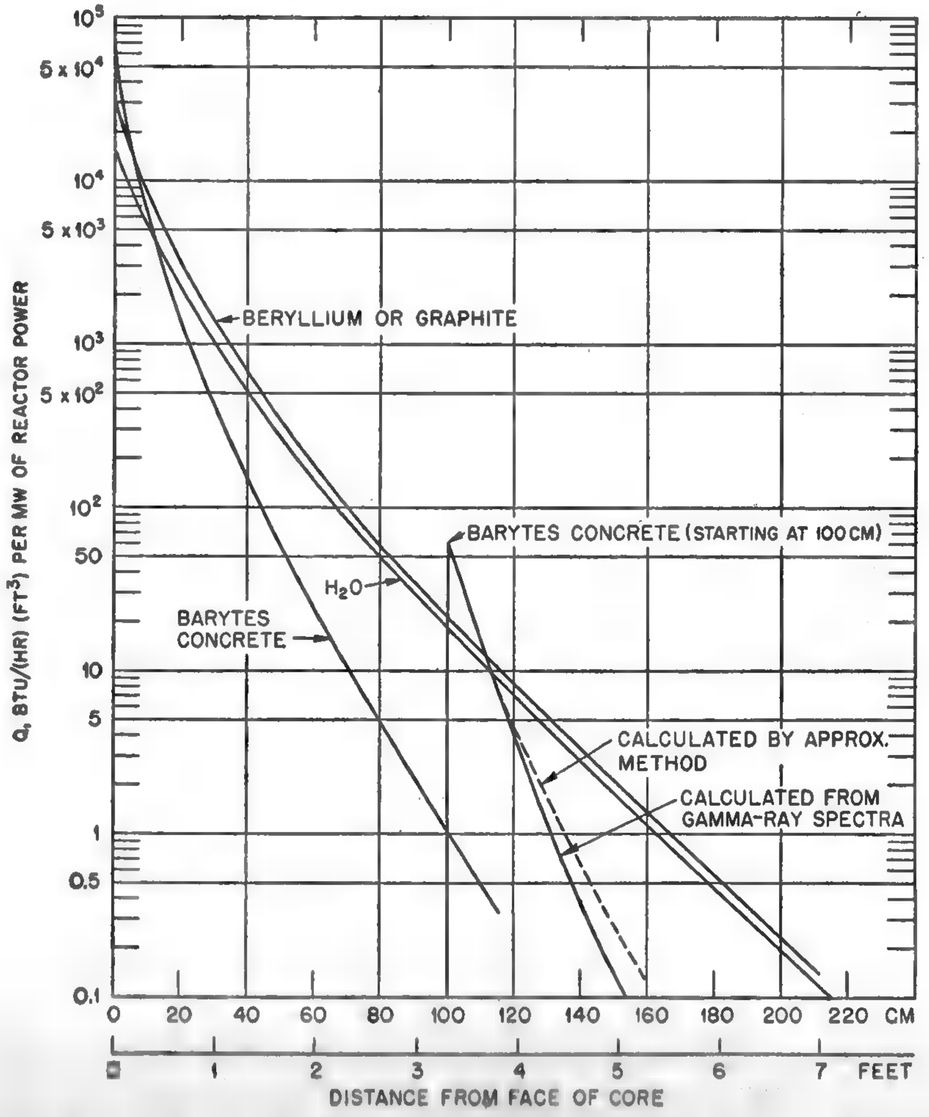


FIG. 11.1 Heat release rate per megawatt of power at various distances from reactor core (J. A. Lane)

11.58. The curves are for reflectors of beryllium, graphite, and water, and also for a shield of barytes concrete in two locations — one at the face of the reactor core and the other at 100 cm from the core with water in between. It will be observed that the volumetric heat-source distribution is essentially the same in beryllium and graphite reflectors because of the approximate equality of the energy-absorption coefficients for gamma rays.

Example Calculate (*a*) the maximum volumetric heat-generation rate at the surface of a barytes concrete shield (density = 3.5 g/cm³) placed 100 cm from a Swimming Pool type reactor core, with water in between, when the operating level is 1 megawatt (Mw); (*b*) the heat-generation rate at a point 2 ft inside the shield.

(*a*) From Fig. 11.1 it is found that the heat-generation rate in water at 100 cm (3.29 ft) from the core face is 17 Btu/(hr)(ft³). In the absence of direct data, the *mass* absorption coefficient, for energy absorption (cf. § 9.43), i.e., μ_e/ρ, of the barytes concrete may be taken to be the same as that for water. Since the density of the concrete is 3.5 times that of water, the (linear) energy-absorption coefficient, μ_e, will be 3.5 times as great. Hence, at the same location as the value in water is 17 Btu/(hr)(ft³), the heat-generation rate will be

$$(3.5)(17) = 60 \text{ Btu/(hr)(ft}^3\text{)}$$

in the concrete.

(*b*) For an accurate calculation of the heat generation inside the shield, it is necessary to know the gamma-ray spectrum at all points concerned, as well as the absorption coefficients for all the energies. An approximate value, however, may be obtained in the following manner.

From the slope of the heat-generation curve in water, the effective linear absorption coefficient in this medium at 100 cm from the reactor face is found to be 0.048 cm^{-1}. The equivalent absorption coefficient in barytes concrete will then be (3.5)(0.048) = 0.168 cm^{-1}. The same effective absorption coefficient in barytes concrete placed directly against the reactor face is found, from the appropriate curve in Fig. 11.1, to occur at a distance of 0.5 ft from the reactor face; at this point the heat generation rate is 2700 Btu/(hr)(ft³). At a further distance of 2 ft into the concrete, i.e., 2.5 ft from the reactor, the heat generation is seen to be 7 Btu/(hr)(ft³), giving a reduction factor of 2.6×10^{-3} for 2 ft of concrete. Applying the same factor to the value of 60 Btu/(hr)(ft³) at the surface, as obtained above, the heat-generation rate at a point 2 ft within the shield, separated from the reactor by 100 cm of water, is found to be $60 \times 2.6 \times 10^{-3} = 0.16$ Btu/(hr)(ft³).

11.59. The dotted curve in Fig. 11.1 shows the heat-generation rates at various points in a barytes concrete shield calculated by the approximate procedure described in the example just given. The calculated values are somewhat higher than those obtained from the more accurate method using the gamma-ray spectrum. However, the discrepancies are not serious except for appreciable distances within the shield, where the heat generation rate is small, in any event.

HEAT TRANSMISSION BY CONDUCTION AND CONVECTION

Introduction

11.60. The purpose of the present section is to consider such aspects of the design of the reactor cooling system as are concerned with the transmission of heat in solid components and the transfer of heat from those components to a coolant across the solid-fluid interface. It will be seen that, as a result, it is possible to determine the temperature distribution in various parts of the reactor. Such a knowledge is of importance since, as stated earlier, a particular temperature may set a limit on the power output. In most cases of heat transmission in a reactor the volumetric heat sources are nonuniform, as seen above; nevertheless, the thermal problems can be treated in a manner analogous to that encountered in conventional practice. It was mentioned in § 11.21 that for purposes of calculation it may sometimes be necessary to idealize the geometry of the system. For this reason, the following treatment will be restricted to simple geometries.

11.61. There are three general mechanisms whereby heat is transferred from one point to another, namely: (1) conduction, (2) convection, and (3) radiation. The last of these usually plays a negligible part in heat removal in reactor systems, and so it will not be considered further here. The discussion will therefore be concerned with heat transmission by conduction and convection.*

Heat Transmission by Conduction

11.62. The term *conduction* refers to the transfer of heat by molecular (and sometimes electronic) interaction without any accompanying macroscopic displacement of matter. The flow of heat by conduction is governed by the Fourier equation

$$q = -kA\frac{dt}{dx}, \tag{11.62.1}$$

where q is the rate (per unit time) at which heat is conducted in the x-direction through a plane of area A normal to this direction, at a point where the temperature gradient is dt/dx.† The quantity, k, defined by equation (11.62.1), is the *thermal conductivity*. The units employed for expressing k must, of course, be consistent with those used for the other variables in equation (11.62.1). Thus, if the heat is given in calories, time in seconds, length in centimeters, and temperature on the Centigrade scale, then the appropriate units of k are cal/(sec)(cm²)(°C/cm), as used in Chapter VIII. On the other hand, if the units used are British thermal units, hours, feet, and degrees Fahrenheit, then k will be in Btu/(hr)(ft²)(°F/ft).‡

* For additional information standard texts on heat transfer should be consulted, e.g., W. H. McAdams, "Heat Transmission," 3rd ed., McGraw-Hill Book Co., Inc., 1954; M. Jakob and G. A. Hawkins, "Elements of Heat Transfer and Insulation," 2nd ed., John Wiley and Sons, Inc., 1950. More detailed treatments are given by L. M. K. Boelter, *et al.*, "Heat Transfer Notes," University of California Press, 1946; M. Jakob, "Heat Transfer," Vol. I, John Wiley and Sons, Inc., 1949. For special applications to reactor systems, see C. Goodman (Ed.), "The Science and Engineering of Nuclear Power," Vol. II, Addison-Wesley Press, Inc., 1949, Chapter 10 (by E. R. Gilliland, *et al.*); C. F. Bonilla, Report M-4476.

† The quantity q/A, having the dimensions of heat/(time)(area), e.g., Btu/(hr)(ft²), is called the heat flux.

‡ For conversion factors, see Appendix, Table A.1.

11.63. The thermal conductivity, k, is a physical property of the medium through which the heat conduction occurs. For anisotropic substances, the value of k is a function of direction; although methods are available for making allowance for such variations, they are ignored in most analytical solutions of conduction problems. The thermal conductivity is also temperature dependent (see Tables A.3, A.4 in Appendix), and can generally be expressed in a power series; thus,

$$k = c_0 + c_1 t + c_2 t^2 + \cdots,$$

which, in many cases, may be approximated to the simple linear form

$$k = k_0(1 + at).$$

Where considerable accuracy is desirable (and possible), allowance must be made for the variation of thermal conductivity with temperature. But very frequently, especially when the temperature range is not great, k is taken to be constant. Some values of interest in reactor design were given in Chapter VIII and others will be found in the Appendix.

11.64. Upon integration of equation (11.62.1), it is found, for unidirectional heat flow by conduction in a slab of constant cross section, with k independent of temperature, that

$$q = -kA \frac{t_1 - t_2}{x_1 - x_2}, \tag{11.64.1}$$

where t_1 and t_2 are the temperatures at two points whose coordinates are x_1 and x_2. The result means that the temperature gradient at a point, i.e., dt/dx, in the Fourier equation (11.62.1) may be replaced by the average gradient over any distance, i.e., $(t_1 - t_2)/(x_1 - x_2)$.

11.65. If $t_1 - t_2$ is replaced by Δt, the temperature difference, and $x_2 - x_1$ by L, the length of the heat-flow path, equation (11.64.1) upon rearrangement takes the form

$$q = \frac{\Delta t}{\frac{L}{kA}}. \tag{11.65.1}$$

This expression is analogous to Ohm's law, $I = E/R$; hence the quantity L/kA is often called the *thermal resistance* for a slab conductor. The analogy between the flow of heat by conduction and of electricity is the basis of the *thermal circuit* concept which is very useful in solving heat-transfer problems. In general, the rate of heat flow, q, is equivalent to the current, I; the temperature difference, Δt, is the analogue of the potential difference (or EMF), E; and the thermal resistance replaces the electrical resistance.

Heat Transmission by Convection

11.66. Heat transmission by *convection*, which is usually concerned with the transfer of heat across a solid-fluid interface, involves macroscopic motion of the fluid. In *free convection* the motion is a consequence of the buoyant forces generated in the fluid due to temperature differences within it. In *forced convection*, on the other hand, the fluid is displaced by mechanical means, e.g., by a pump. When applied to heat removal in reactors, two aspects of convective heat transmission must be considered; there is, first, transfer of heat from the material which is being cooled, e.g., the fuel element, to the coolant; and, second, the transport of this heat, usually in sensible form, by flow of the coolant from one point in the system to another. Fluid flow is thus an important problem, as will be seen later, in the study of reactor cooling systems.

11.67. The fundamental equation of convective heat transfer, for both free and forced motion of the fluid, is Newton's law of cooling, which may be written as

$$q = hA_h \Delta t, \tag{11.67.1}$$

where q is the rate of convective heat transfer to or from a surface of area A_h when the temperature difference is Δt. The quantity h, defined by equation (11.67.1), is usually called the *heat-transfer coefficient*, but some authors prefer the term *unit thermal conductance*. For engineering calculations, it is usually expressed in units of Btu/(hr)(ft²)(°F). It should be noted that equation (11.67.1) applies to convective heat-flow in either direction, i.e., from solid to fluid or from fluid to solid; the actual direction of the flow depends, of course, upon the sign of Δt.

11.68. Although the value of h is dependent upon the physical properties of the fluid medium, it is also a function of the shape and dimensions of the interface, and of the nature, direction, and velocity of the fluid flow. Thus, the heat-transfer coefficient is a property of the particular system under consideration. Another factor which determines h is the exact definition of Δt, i.e., the temperature difference between the surface and the fluid. Whereas the temperature of the solid is uniquely defined, that of the fluid is subject to several arbitrary definitions. The latter is usually taken as the "mixed-mean fluid temperature," t_f, given by

$$t_f \equiv \frac{\int_{A_f} \rho c_p v t dA_f}{\int_{A_f} \rho c_p v dA_f},$$

where ρ, c_p, and v are the density, specific heat, and flow velocity, respectively, of the fluid, and A_f is the cross-sectional (or transverse) flow area of the fluid. The value of the heat-transfer coefficient in any given circumstances can be determined experimentally, but for design purposes the general practice is to use results predicted by means of various theoretical and semiempirical expressions (see § 11.124, *et seq.*).

11.69. By slight rearrangement, equation (11.67.1) can be written in the Ohm's law form

$$q = \frac{\Delta t}{\dfrac{1}{hA_h}},$$

so that, as before, q and Δt are analogous to current and potential difference, respectively, but $1/hA_h$ now represents the thermal resistance to convective heat transfer. Thermal circuits can thus be constructed involving one or more stages of thermal conduction combined with convection for the solution of problems involving both types of heat transfer, as will be shown in the succeeding paragraphs.

Heat Conduction in Infinite Slab with Convection Boundary Conditions

11.70. Although thermal problems in reactors generally involve internal sources, the first (and simplest) case to be considered is that of an infinite slab of finite thickness with *no internal heat source.* It is postulated that heat is transferred at both faces by convection and that the heat-transfer coefficients are h_a and h_b, respectively. The idealization of an infinite slab is a good approximation to real (finite) systems in which the transverse dimension is large compared with the thickness, e.g., the cladding of a parallel-plate type fuel element, or the flat plate of a heat exchanger. In the following treatment, steady-state conditions with the thermal conductance independent of temperature will be postulated.

11.71. The characteristics of the physical system are shown at the left in Fig. 11.2, and the equivalent thermal-circuit diagram is given at the right. The total thermal resistance is the sum of three terms; two (R_1 and R_3) are for the convective resistances and the third (R_2) is for the conductive resistance of

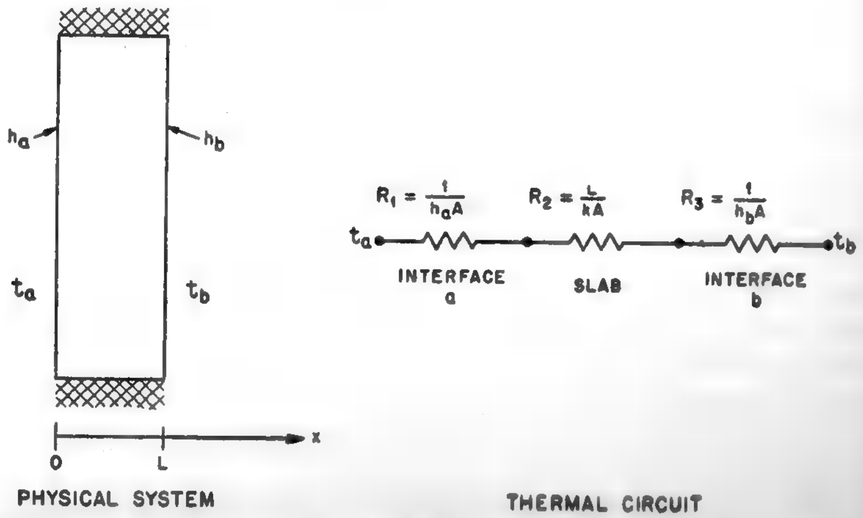


Fig. 11.2 Heat conduction in infinite slab with convection boundaries

the slab; hence, if A is the heat-flow area, the total thermal resistance, R, is

$$R = \frac{1}{h_aA} + \frac{L}{kA} + \frac{1}{h_bA} = \frac{1}{UA}. \tag{11.71.1}$$

The relationship between the heat-flow rate and the temperature difference $t_a - t_b$, which is equal to Δt, is thus given by

$$q = \frac{t_a - t_b}{R} = UA(t_a - t_b)$$

or

$$t_a - t_b = \frac{q}{UA}.$$

The quantity U, defined by equation (11.71.1), is called the *over-all coefficient of heat transfer;* it has the same dimensions as the heat-transfer coefficient.

11.72. The equations derived above may be used to determine the steady-state difference in temperature, $t_a - t_b$, between two fluids in a heat exchanger, if the heat-flow rate, q, is given. Alternatively, if $t_a - t_b$ is fixed, the corresponding value of q can be calculated for specified conditions. The individual temperature differences, i.e., the two fluid-solid differences and that between the two faces of the slab, can be determined by the use of equations (11.67.1) and (11.64.1), respectively.

Heat Conduction in Hollow Cylinder with Convection Boundary Conditions

11.73. Tubular heat-transfer surfaces are used in many heat exchangers, and the performance of these can be analyzed on the basis of heat conduction through hollow circular cylinders with convection at both interior and exterior surfaces. The physical system in cross section and the thermal-circuit diagram are shown in Fig. 11.3; it is assumed that all the heat flow is in a radial direction. The inner and outer radii of the cylinder are a and b, respectively; h_a and h_b are the

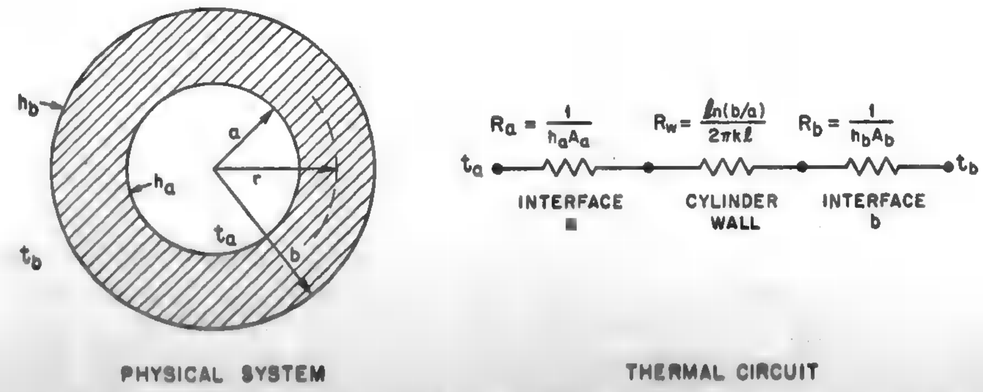


Fig. 11.3 Heat conduction in hollow cylinder with convection boundaries

heat-transfer coefficients for the inner and outer fluids, and t_a and t_b are the mixed-mean temperatures of inner and outer fluids, respectively.

11.74. The convective thermal resistances are equal to $1/h_aA_a$ and $1/h_bA_b$, respectively, where A_a and A_b are the areas of the inner and outer surfaces, respectively. The thermal conductive resistance of the cylinder wall remains to be determined. If l is the length of the cylinder, then the heat-flow rate, expressed at an arbitrary radius r, due to conduction within the wall is given by equation (11.62.1) as

$$q = -k(2\pi rl)\frac{dt}{dr},$$

where k is the thermal conductivity and $2\pi rl$ is equivalent to A, the area through which heat flow occurs. This expression may be rearranged and integrated over the wall thickness, i.e., from $r = a$ to $r = b$; the result is

$$q = \frac{\Delta t}{\dfrac{\ln (b/a)}{2\pi kl}}, \tag{11.74.1}$$

where Δt is the difference in temperature across the cylinder wall.

11.75. It is evident from equation (11.74.1) that the thermal resistance, R_w, of the wall is given by

$$R_w = \frac{\ln (b/a)}{2\pi kl}. \tag{11.75.1}$$

The total thermal resistance of the system under consideration is consequently

$$R = \frac{1}{h_aA_a} + \frac{\ln (b/a)}{2\pi kl} + \frac{1}{h_bA_b} = \frac{1}{U_bA_b},$$

where U_b is the over-all heat-transfer coefficient based on the *outer surface* of the cylinder. According to the thermal-circuit concept

$$q = \frac{t_a - t_b}{R} = U_bA_b(t_a - t_b)$$

or

$$t_a - t_b = \frac{q}{U_bA_b},$$

where

$$U_b = \frac{1}{\dfrac{b}{h_aa} + \dfrac{b \ln (b/a)}{k} + \dfrac{1}{h_b}}. \tag{11.75.2}$$

An example of the application of these results will be given later (§ 11.185).

HEAT TRANSMISSION IN SYSTEMS WITH INTERNAL SOURCES *

Generalization from One-dimensional Slab

11.76. The treatment of heat transmission in the steady state will now be extended to systems with internal sources and, for this purpose, the simple, one-

* See L. M. K. Boelter, *et al.*, *op. cit.*, Chapter VII; M. Jakob, *op. cit.*, Chapter 10.

dimensional case of an infinite slab will be examined (Fig. 11.4). Consider a thin section of the slab of thickness dx at x; then in the steady state the heat rate balance requires that

Heat conducted out of $A\,dx$ − Heat conducted into $A\,dx$ = Heat generated in $A\,dx$,

where A is the heat-conduction area, so that $A\,dx$ is the volume of the section. By equation (11.62.1)

$$q_{\text{in}} = -kA\left(\frac{dt}{dx}\right)_x$$

and

$$q_{\text{out}} = -kA\left(\frac{dt}{dx}\right)_{x+dx},$$

assuming the heat flow is from left to right. Taking the difference of these terms and expanding the expression for q_{out}, it is found that

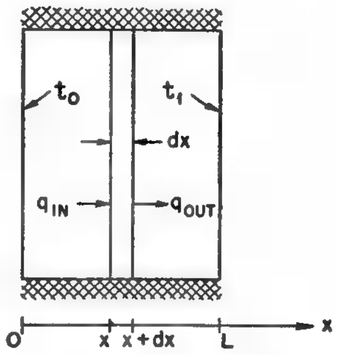


Fig. 11.4 Heat transmission in infinite slab with internal source

$$q_{\text{out}} - q_{\text{in}} = -kA\left\{\left[\left(\frac{dt}{dx}\right)_x + \left(\frac{d^2t}{dx^2}\right)_x dx + \cdots\right] - \left(\frac{dt}{dx}\right)_x\right\}$$
$$= -kA\left(\frac{d^2t}{dx^2}\right)dx, \tag{11.76.1}$$

higher order terms in the expression being neglected.

11.77. If $Q(x)$ represents the volumetric thermal source strength, expressed as heat generated per unit time per unit volume at x, e.g., in Btu/(hr)(ft^3), then $Q(x)A\,dx$ is the heat generated per unit time in the section of the slab under consideration. It follows, therefore, from the heat-balance equation and from equation (11.76.1) that

$$-kA\left(\frac{d^2t}{dx^2}\right)dx = Q(x)A\,dx$$

so that

$$-k\frac{d^2t}{dx^2} = Q(x), \tag{11.77.1}$$

where $Q(x)$ may or may not vary with x.

11.78. It can be shown that equation (11.77.1) is a particular form of the general equation for the steady state in a system with an internal source, namely,

$$-k\nabla^2t = Q(\mathbf{r}), \tag{11.78.1}$$

where the thermal conductivity k is the same in all directions, i.e., for a truly isotropic medium, and ∇^2 is the Laplacian operator (§ 3.16). It is apparent that in the one-dimensional case, in Cartesian coordinates, equation (11.78.1) reduces to equation (11.77.1). For conduction in a cylinder, in the radial direction, equation (11.78.1) becomes

$$-k\left(\frac{d^2t}{dr^2}+\frac{1}{r}\cdot\frac{dt}{dr}\right)=Q(r) \qquad (11.78.2)$$

and for a sphere

$$-k\left(\frac{d^2t}{dr^2}+\frac{2}{r}\cdot\frac{dt}{dr}\right)=Q(r). \qquad (11.78.3)$$

Conduction in Infinite Slab with Uniform Source

11.79. Many reactor designs, e.g., MTR, Swimming Pool type, etc., have plate-type fuel elements which can be idealized as an infinite slab with a uniformly distributed thermal source. In this case, equation (11.77.1) may be used with Q constant, independent of x. The general solution of this differential equation is then

$$t=-\frac{Qx^2}{2k}+C_1x+C_2, \qquad (11.79.1)$$

where the constants C_1 and C_2 are to be evaluated from the boundary conditions, which in this case (see Fig. 11.4) are

$$x=0,\ t=t_0 \quad \text{and} \quad x=L,\ t=t_1.$$

It is thus found that

$$t=-\frac{Qx^2}{2k}+\left(\frac{t_1-t_0}{L}+\frac{QL}{2k}\right)x+t_0, \qquad (11.79.2)$$

which gives the steady-state temperature t at any point x in the slab. Differentiating equation (11.79.2) with respect to x to obtain dt/dx, and inserting the result in equation (11.62.1), it follows that

$$\frac{q}{A}=\frac{Q}{2}(2x-L)+\frac{k(t_0-t_1)}{L}. \qquad (11.79.3)$$

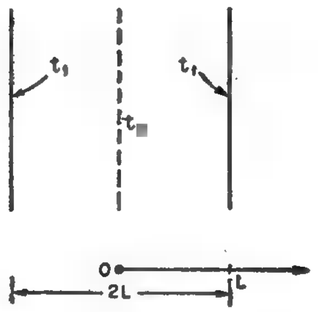


Fig. 11.5 Temperatures in plate-type fuel element

11.80. The slab of thickness L shown in Fig. 11.4 may be regarded as the right half of a plate-type fuel element, of thickness $2L$, in which both surface temperatures are the same. In this case $q=0$ at $x=0$, at the middle of the fuel element, since the temperature is a maximum and dt/dx is, consequently, zero at this point. If q is equal to zero for $x=0$, equation (11.79.3) reduces to

$$t_0-t_1=\frac{QL^2}{2k},$$

where t_0-t_1 represents the temperature difference from the central plane to the outer surface of the fuel element (Fig. 11.5).

Heat Transmission in Clad Plate-type Fuel Element

11.81. The physical system for a plate-type fuel element clad on each face to form a "sandwich" is shown in Fig. 11.6. Here a is the half-thickness of the fuel portion and b is the half-thickness of the sandwich; t_0 is the temperature at the central plane of the fuel and t_1 is that at the fuel-cladding interface. The flow of heat is assumed to occur only in the x-direction.

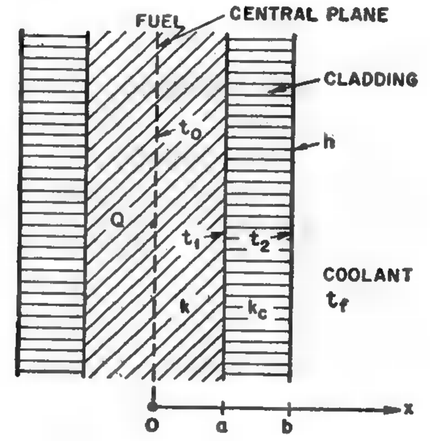


Fig. 11.6 Heat transmission in clad plate-type fuel element

11.82. The internal heat source is in the fuel-bearing region of the element, and here equation (11.77.1) is applicable, i.e.,

$$-k\frac{d^2t}{dx^2}=Q, \qquad 0\leqslant x\leqslant a,$$

and if the heat source is assumed to be uniform, the general solution is given by equation (11.79.1). The boundary conditions are

$$\frac{dt}{dx}=0,\ x=0 \quad \text{and} \quad t=t_1,\ x=a,$$

so that the appropriate solution is found to be

$$t-t_1=\frac{Q}{2k}(a^2-x^2), \qquad 0\leqslant x\leqslant a. \qquad (11.82.1)$$

By means of this equation, the steady-state temperature t at any point x, in the fuel-bearing region, can be calculated. The temperature distribution in the fuel element may be assumed to be symmetrical about the central plane, so that equation (11.82.1) also applies to negative values of x.

11.83. The heat transmission through the cladding into the coolant fluid can now be treated by the simple thermal-circuit method, with convection at one boundary;* thus, if t_f is the mixed-mean temperature of the fluid, then

$$\begin{aligned}t_1-t_f&=q(R_{\text{clad}}+R_{\text{conv}})\\&=\frac{q}{A}\left(\frac{b-a}{k_c}+\frac{1}{h}\right),\end{aligned} \qquad (11.83.1)$$

where $R_{\text{clad}}=(b-a)/k_cA$ is the thermal resistance of the cladding (thermal conductivity $=k_c$) and $R_{\text{conv}}=1/hA$ is the convective thermal resistance at

* The small quantity of heat liberated in the cladding from various sources is neglected in this calculation.

the cladding-fluid interface (heat-transfer coefficient = h); A is the area of the heat-transfer surface. From a heat-rate balance, q at the interface is found to be AQa. Since there is no thermal source in the cladding, this is the value of q in equation (11.83.1); hence

$$t_1 - t_f = Qa\left(\frac{b-a}{k_c} + \frac{1}{h}\right). \tag{11.83.2}$$

This gives the difference in temperature between the fuel-cladding interface and the coolant.

11.84. For any point x in the cladding, the corresponding temperature difference $t - t_f$ is obtained by substituting x for a in the thermal resistance term in equation (11.83.2); the result is

$$t - t_f = Qa\left(\frac{b-x}{k_c} + \frac{1}{h}\right), \qquad a \leqslant x \leqslant b. \tag{11.84.1}$$

At the outside surface of the cladding, where $x = b$, the temperature t_2 is given simply by

$$t_2 - t_f = \frac{Qa}{h}. \tag{11.84.2}$$

11.85. If there is no thermal resistance at the surface of contact between the fuel-bearing region and the cladding (cf. § 11.98), the difference in temperature between any point in the fuel portion and the coolant can be obtained by adding equations (11.82.1) and (11.83.2); thus,

$$t - t_f = \frac{Q}{2k}(a^2 - x^2) + Qa\left(\frac{b-a}{k_c} + \frac{1}{h}\right), \qquad 0 \leqslant x \leqslant a. \tag{11.85.1}$$

At the center of the fuel element $x = 0$ and $t = t_0$; hence,

$$t_0 - t_f = \frac{Qa^2}{2k} + Qa\left(\frac{b-a}{k_c} + \frac{1}{h}\right). \tag{11.85.2}$$

This gives the maximum temperature difference in the system.

Example Determine the steady-state temperature distribution in a plate-type fuel element consisting of 1 mm of a uranium-aluminum alloy, containing 18 per cent by weight of uranium enriched to 90 per cent in U-235, clad on each side with 0.5 mm thickness of aluminum. The thermal conductivities of the fuel region and the cladding may be taken to be the same, namely, 118 Btu/(hr)(ft²)(°F/ft). The thermal-neutron flux in the fuel region is 10^{14} neutrons/(cm²)(sec). The plate is cooled on both sides by water whose temperature is 80°F, and the heat-transfer coefficient for the given conditions is 7000 Btu/(hr)(ft²)(°F). (The thermal source in the fuel-bearing section may be assumed to be uniform and k may be taken as being independent of temperature.)

The first problem is to determine the thermal source strength, Q; this is equal to $\Sigma_f \phi E_f$ where Σ_f is the thermal macroscopic fission cross section in the fuel region, ϕ is the thermal-neutron flux (which is known), and E_f is the energy released in the fuel per fission (which may be taken as 200 Mev). Hence it is required to evaluate $\Sigma_f = N\sigma_f$, where σ_f is the thermal fission cross section of uranium-235 (about 520×10^{-24} cm² at ~120°F) and N is the number of these nuclei per cm³ of the fuel.

Taking the density of uranium as 19 g/cm³ and that of aluminum as 2.7 g/cm³, the density of the fuel region (18% U–82% Al by weight) is found to be 3.2 g/cm³. Since the uranium contains 90% of U-235, there are (0.9)(0.18) g of U-235 per gram of fuel, or (0.9)(0.18)(3.2) g of U-235 per cm³ of fuel. Each 235 g of U-235 contains 6.02×10^{23} (the Avogadro number) nuclei, so that

$$\Sigma_f = N\sigma_f = \frac{(0.9)(0.18)(3.2)(6.02 \times 10^{23})(520 \times 10^{-24})}{235}$$
$$= 0.69 \text{ cm}^{-1}.$$

The fission rate is $\Sigma_f\phi = 0.69 \times 10^{14}$ fissions/(sec)(cm³), and so, at 200 Mev/fission, the rate of energy release is 1.38×10^{16} Mev/(sec)(cm³); hence,

$$Q = 1.38 \times 10^{16} \frac{\text{Mev}}{(\text{sec})(\text{cm}^3)} \times 1.52 \times 10^{-16} \frac{\text{Btu}}{\text{Mev}} \times 3600 \frac{\text{sec}}{\text{hr}} \times 2.83 \times 10^4 \frac{\text{cm}^3}{\text{ft}^3}$$
$$= 2.1 \times 10^8 \text{ Btu/(hr)(ft}^3\text{)}.$$

Noting that $a = 0.05$ cm $= (0.05)(3.28 \times 10^{-2}) = 1.64 \times 10^{-3}$ ft, $b = 0.1$ cm $= 3.28 \times 10^{-3}$ ft, $t_f = 80$°F, $k = k_c = 118$ Btu/(hr)(ft²)(°F/ft), and $h = 7800$ Btu/(hr)(ft²)(°F), the temperature t_0, at the central plane, where $x = 0$, is given by equation (11.85.2) as 131.4°F. Similarly, from equation (11.83.2), t_1 is found to be 129°F and,

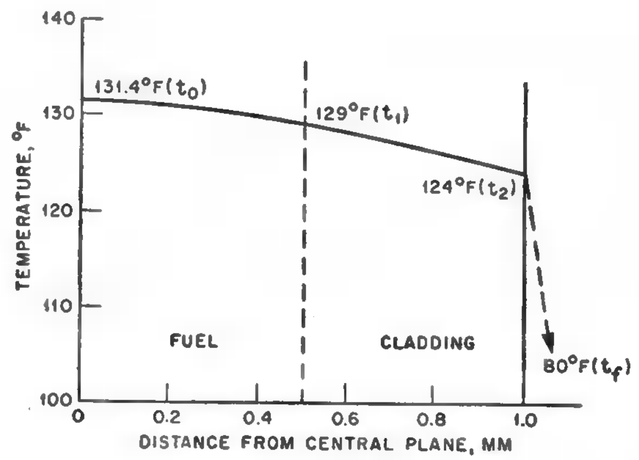


FIG. 11.7 Temperature distribution in clad plate-type fuel element

from equation (11.84.2), the value of t_2 is 124°F. Temperatures between t_0 and t_1 are found from equation (11.82.1) and from t_1 to t_2 from equation (11.84.1); the results are plotted in Fig. 11.7. The dotted portion indicates that the temperature distribution in the fluid is not determined here.

HEAT TRANSMISSION IN CLAD CYLINDRICAL FUEL ELEMENT

11.86. In many reactors, e.g., X-10, Brookhaven, NRX, etc., the fuel elements are in the form of cylindrical slugs, usually clad with aluminum, the heat being removed from the outer surface by convection. The determination of the steady-state temperature distribution is similar to that described for a

plate-type fuel element. Consider first the fuel cylinder of radius a (Fig. 11.8), with uniform source Q. For heat conduction in the radial direction, equation (11.78.2) is applicable, the general solution being

$$t = -\frac{Qr^2}{4k} + C_1 \ln r + C_2, \qquad 0 \leqslant r \leqslant a. \tag{11.86.1}$$

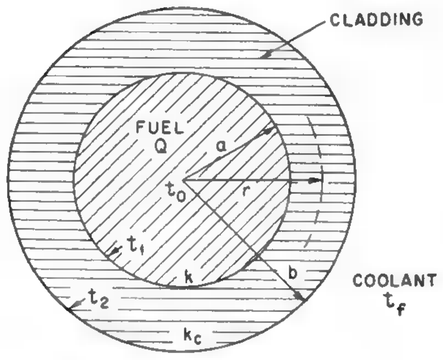


FIG. 11.8 Heat transmission in clad cylindrical fuel element

The boundary conditions in the present case are

$$\frac{dt}{dr} = 0, r = 0 \quad \text{and} \quad t = t_1, r = a,$$

so that the appropriate solution is

$$t - t_1 = \frac{Q}{4k}(a^2 - r^2), \qquad 0 \leqslant r \leqslant a,$$

where t is the temperature at the radial distance r. If t_0 is the temperature along the central axis, where $r = 0$, then

$$t_0 - t_1 = \frac{Qa^2}{4k} \tag{11.86.2}$$

for the temperature drop across the fuel itself.

11.87. For the transmission of heat through the cladding to the coolant,

$$t_1 - t_f = q(R_{\text{clad}} + R_{\text{conv}})$$

as before, where R_{clad} is identical with R_w of § 11.75 and is given by equation (11.75.1), and R_{conv} is $1/hA$, where A is the outside area of the cylinder of length l, i.e., $2\pi bl$. It follows, therefore, that

$$t_1 - t_f = \frac{q}{2\pi l}\left(\frac{1}{k_c}\ln\frac{b}{a} + \frac{1}{hb}\right).$$

At the interface between fuel and cladding, where $x = a$, $dt/dr = -Qa/2k$; since the surface area is here $2\pi al$, it follows from a heat-rate balance that

$$q = \pi a^2 l Q$$

and, hence,

$$t_1 - t_f = \frac{Qa^2}{2}\left(\frac{1}{k_c}\ln\frac{b}{a} + \frac{1}{hb}\right). \tag{11.87.1}$$

This gives the temperature difference between the fuel-cladding interface and the coolant.

11.88. Upon adding equations (11.86.2) and (11.87.1), the temperature drop from the center of the fuel element to the coolant is found to be

$$t_0 - t_f = \frac{Qa^2}{4k} + \frac{Qa^2}{2}\left(\frac{1}{k_c}\ln\frac{b}{a} + \frac{1}{hb}\right), \tag{11.88.1}$$

assuming negligible thermal resistance at the fuel-cladding interface. Equations for the temperature distribution in the cladding or for the difference between the temperature at any point in the fuel or the cladding and the coolant can be readily derived, if required, by a procedure similar to that used for plate-type elements.

Example Determine the maximum specific power (in kw/kg) obtainable with a water-cooled, cylindrical fuel rod of natural uranium, 0.9 in. in diameter, having an aluminum cladding 50 mils (0.05 in.) thick, assuming that the maximum metal temperature does not exceed 1000°F and the maximum mean coolant temperature does not exceed 200°F. What is the temperature drop through (*a*) the fuel, (*b*) the cladding, (*c*) between the cladding and coolant, at maximum power? The following data are to be used: For uranium, $k = 14$ and for aluminum, $k = 118$ Btu/(hr)(ft²)(°F/ft); $h = 6000$ Btu/(hr)(ft²)(°F), a representative value for water flowing at about 30 ft/sec in a narrow channel.

The specific power is related to Q, the volumetric source strength, since this is a measure of the rate of heat release by fission per unit volume of fuel. Hence, the maximum value of Q must be determined; this can be done by means of equation (11.88.1), setting $t_0 = 1000$°F, and $t_f = 200$°F, so as to give the maximum permissible temperature drop. Then, with $a = 0.45$ in. $= 0.0375$ ft, and $b = 0.50$ in. $= 0.0416$ ft,

$$1000 - 200 = \frac{Q(0.0375)^2}{(4)(14)} + \frac{Q(0.0375)^2}{2}\left[\frac{1}{118}\ln\frac{0.50}{0.45} + \frac{1}{(6000)(0.0416)}\right]$$

$$= 2.85 \times 10^{-5}Q$$

$$Q = \frac{800}{2.85 \times 10^{-5}} = 2.80 \times 10^7 \text{ Btu/(hr)(ft}^3\text{)}.$$

Since 1 Btu/hr $= 2.93 \times 10^{-4}$ kw, $Q = 8.22 \times 10^3$ kw/ft³. The density of uranium is about 19 g/cm³, i.e., 0.019 kg/cm³, so that 1 ft³ $= 2.83 \times 10^4$ cm³ of uranium weighs $(2.83 \times 10^4)(0.019) = 5.37 \times 10^2$ kg. The required maximum specific power is consequently $8.22 \times 10^3/5.37 \times 10^2 = 15.4$ kw/kg of fuel. The specific power per kilogram of fissionable material is 140 times this value, i.e., $(140)(1.45) = 2160$ kw/kg of U-235.

The temperature drop across the fuel is $t_0 - t_1$, and by equation (11.86.2), using Btu as the heat unit in Q and k, this is

$$t_0 - t_1 = \frac{Qa^2}{4k} = \frac{(2.80 \times 10^7)(0.0375)^2}{(4)(14)}$$

$$= 704\text{°F}.$$

The temperature drop across the cladding is given by the first term in the parentheses in equation (11.88.1); hence,

$$t_1 - t_2 = \frac{Qa^2}{2k_c}\ln\frac{b}{a} = \frac{(2.80 \times 10^7)(0.0375)^2}{(2)(118)}\ln\frac{0.50}{0.45}$$

$$= 17\text{°F}.$$

The temperature drop between the cladding and coolant is obtained from the second term in the parentheses in equation (11.88.1); thus,

$$t_2 - t_f = \frac{Qa^2}{2hb} = \frac{(2.80 \times 10^7)(0.0375)^2}{(2)(6000)(0.0416)}$$

$$= 79\text{°F}.$$

(It will be noted that the three temperatures add up to 800°F, as they should.)

Heat Transmission in Hollow Cylindrical Fuel Element

11.89. There has been some interest in a hollow, cylindrical fuel element, cooled internally and externally, in order to increase the heat-transfer surface. This case will be considered briefly. The steady-state temperature distribution in the fuel region is given by the differential equation (11.78.2), of which the general solution is equation (11.86.1), assuming a uniform source and radial heat transmission.

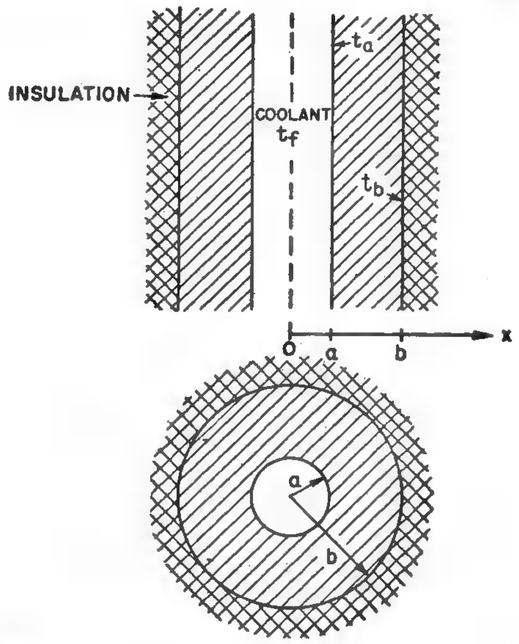


Fig. 11.9 Heat transmission in insulated cylinder

11.90. An appropriate solution can be obtained for a long cylinder in which the steady-state temperature distribution has become established, the boundary conditions being that the heat conducted out of each surface is convected away by the corresponding coolant stream. The mixed-mean coolant temperature for each side is obtained by making a heat-rate balance on the system, i.e., the total heat generated in the fuel element is lost by convection to the two coolant streams.

11.91. The procedure for determining the temperature distribution in the internal and external cladding, using the thermal-circuit concept, is similar to that described in preceding sections and need not be given here. An accurate analysis of the temperature distribution in the fuel requires, of course, that the cladding be taken into consideration in the basic equations.

Heat Transmission in Moderator: Insulated Cylinder with Convection

11.92. A special case of heat transmission in a hollow cylinder arises in the treatment of the temperature distribution in a moderator structure pierced with regularly spaced coolant channels. It is a good idealization then to assume that the region surrounding each coolant channel constitutes an insulated cylinder with a uniform heat source and an internal convective boundary (see Fig. 11.9). The treatment is applicable also to an internally cooled cylindrical fuel element surrounded by moderator.

11.93. As before, the general solution is given by equation (11.86.1), the constants C_1 and C_2 being obtained from the following boundary conditions. Since all the heat flow is toward the center of the channel, it is apparent that

a maximum temperature will occur at the outer surface of the insulated cylinder, where $r = b$. This leads to the boundary condition

$$\left(\frac{dt}{dr}\right)_{r=b} = 0,$$

and the other condition is

$$t = t_a, \qquad r = a.$$

It can then be readily shown that

$$t - t_a = -\frac{Q}{4k}(r^2 - a^2) + \frac{Qb^2}{2k}\ln\frac{r}{a}, \tag{11.93.1}$$

which gives the temperature distribution within the moderator.

11.94. In order to determine the distribution with respect to the coolant temperature, use is made of the fact that, in the steady state, all heat reaching the moderator-coolant interface (where $r = a$) by conduction is transmitted to the coolant by convection; hence,

$$k\left(\frac{dt}{dr}\right)_{r=a} = h(t_a - t_f).$$

Utilization of this result together with the expression for dt/dr at $r = a$, obtained by differentiation of equation (11.93.1), gives

$$t_a - t_f = \frac{Q}{2h}\left(\frac{b^2}{a} - a\right),$$

for the temperature drop from the inner surface to the fluid. Hence,

$$t - t_f = \frac{Q}{2k}\left(\frac{a^2 - r^2}{2} + b^2\ln\frac{r}{a}\right) + \frac{Q}{2h}\left(\frac{b^2}{a} - a\right)$$

and

$$t_b - t_f = \frac{Q}{2k}\left[\frac{a^2}{2} + b^2\left(\ln\frac{b}{a} - \frac{1}{2}\right)\right] + \frac{Q}{2h}\left(\frac{b^2}{a} - a\right),$$

where t_b is the maximum temperature attained in the moderator.

Heat Transmission in Shields and Pressure Vessels: Slab with Exponential Heat Source

11.95. In all the preceding examples it has been postulated for simplicity, since it is a good approximation, that the heat source is uniformly distributed within a conductor. A situation will now be considered in which the heat source has an exponential distribution. This is often the case in external reactor components, such as shields and thick-walled pressure vessels, where the heat is largely due to the absorption of gamma radiation, so that the heat source falls off exponentially. When the shield (or pressure vessel) is a sphere or cylinder of large dimensions or rectangular in shape, it may be treated as a slab.

11.96. In these circumstances, the volumetric heat source is represented by $Q = Q_0 e^{-\mu_e x}$ where μ_e is the energy-absorption coefficient (§ 9.43); equation (11.77.1) then takes the form

$$-k\frac{d^2t}{dx^2} = Q_0 e^{-\mu_e x},$$

of which the general solution, provided k is constant, is

$$t = -\frac{Q_0}{k\mu_e^2}e^{-\mu_e x} + C_1 x + C_2.$$

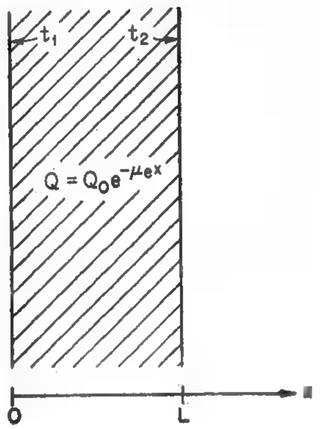


FIG. 11.10 Heat transmission in slab with exponential source

The boundary conditions are (see Fig. 11.10),

$$t = t_1,\ x = 0 \quad \text{and} \quad t = t_2,\ x = L,$$

where L is the thickness of the slab. These lead to the solution

$$t - t_1 = (t_2 - t_1)\frac{x}{L} + \frac{Q_0}{k\mu_e^2}\left[(e^{-\mu_e L} - 1)\frac{x}{L} - e^{-\mu_e x} + 1\right] \qquad (11.96.1)$$

for the steady-state temperature distribution in the slab. It may be noted that the first term on the right represents the linear temperature distribution due only to the difference in temperature between the two faces of the slab, whereas the second gives the effect of the exponential heat source.

11.97. Under certain conditions the combination of the two terms just mentioned leads to a temperature maximum within the slab. The point at which this occurs may be obtained by equating to zero the derivative of equation (11.96.1) with respect to x. The result is

$$(x)_{t_{\max}} = -\frac{1}{\mu_e}\ln\left[(t_1 - t_2)\frac{k\mu_e}{Q_0 L} + \frac{1 - e^{-\mu_e L}}{\mu_e L}\right]. \qquad (11.97.1)$$

The maximum temperature can be obtained by inserting this value for x into equation (11.96.1).

Example A water-cooled and -moderated power reactor is contained within a thick-walled pressure vessel. This vessel is protected from excessive irradiation (and, thus, excessive thermal stress) by a series of steel thermal shields between the reactor core and the pressure vessel. One of these shields, 2 in. thick, whose surfaces are both maintained at 500°F, receives a gamma-ray energy flux of 10^{14} Mev/(cm²)(sec). Calculate the location and magnitude of the maximum temperature in this shield. The energy-absorption coefficient of the radiation in the steel may be taken to be 0.164 cm^{-1}, and the thermal conductivity as 23 Btu/(hr)(ft²)(°F/ft).

In this case, t_1 and t_2 are both 500°F, so that $t_1 - t_2$ is zero. The thickness of the shield, i.e., L (Fig. 11.10), is 2 in. = 1/6 ft, and μ_e in ft^{-1} is given by

$$\mu_e = (0.164)(30.5) = 5.0 \text{ ft}^{-1}.$$

Hence, by equation (11.97.1),

$$(x)_{t_{\max}} = -\frac{1}{5.0}\ln\left(\frac{1 - e^{-5}}{\frac{5}{6}}\right)$$
$$= 0.078 \text{ ft (or 0.94 in.)}.$$

The maximum temperature is thus attained just inside the midplane of the shield.

The maximum temperature is calculated from equation (11.96.1) with $t_1 = 500$°F, $k = 23$ Btu/(hr)(ft²)(°F/ft), and $x = 0.078$ ft. The value of Q_0 is given in § 9.44 as $\phi E \mu_e$; since ϕE is here 10^{14} Mev/(cm²)(sec) and μ_e is 0.164 cm^{-1}, then

$$Q_0 = (10^{14})(0.164) = 1.64 \times 10^{13} \text{ Mev/(cm}^3\text{)(sec)}$$
$$= 2.54 \times 10^5 \text{ Btu/(ft}^3\text{)(hr)},$$

so that

$$t_{\max} - 500 = \frac{(2.54 \times 10^5)}{(23)(5.0)^2}\left[(e^{-5} - 1)\frac{(0.078)}{\frac{1}{6}} - e^{-(5)(0.078)} + 1\right]$$
$$= 25\text{°F}$$

and $t_{\max}$ is consequently 525°F.

CONTACT RESISTANCES BETWEEN SOLIDS

11.98. In the calculation given above for clad fuel elements, it was assumed that the thermal contact resistance at the fuel-cladding interface was negligible because there is usually metallurgical bonding. The resistance to the flow of heat between two solid surfaces in simple mechanical contact is highly unpredictable. An estimate can sometimes be made concerning the effectiveness of a given contact. For example, a pessimistic estimate can be obtained by calculating the resistance across a gas film of the thickness indicated by the roughness of the surfaces. Increasing the contact pressure will reduce the thermal resistance because of minute deformation of the asperities.

11.99. As the theoretical estimates are uncertain, as much experimental data as possible should be obtained from a mocked-up system. Thermal conductances of the order of 10,000 to 50,000 Btu/(hr)(ft²)(°F) have been observed where good contact is maintained.

11.100. Thermal cycling, i.e., alternate increase and decrease of temperature, may misalign surfaces which had previously been in reasonably good contact, and thus decrease the conductance. For example, if a cylindrical cladding jacket is drawn on to a fuel slug (or element) by passing it through a die, the inner surface of the jacket will be pressed into irregularities of the slug surface. Heating of this assembly, particularly from the inside, e.g., by fission, may stretch the jacket so that upon cooling it fits rather loosely, and subsequent reheating may not result in perfect alignment between the fuel element and its cladding.

11.101. In some reactor systems, the heat flow from a fuel element in contact with a solid moderator is an important factor in cooling the fuel element. For example, in some air-cooled natural uranium-graphite reactors, e.g., the Oak

Ridge X-10 reactor, the cylindrical fuel element rests on the graphite moderator in an air channel. The heat flow from the fuel element to the graphite and then to the air passing through the channel helps to lower the temperature of the fuel. Heat-transfer measurements with a solid, cylindrical fuel element resting in one corner of a square channel in the graphite, in a mock-up of an X-10 fuel slug and channel (Fig. 11.11), gave a value of 2.5 Btu/(hr)(ft)(°F) for the uranium to graphite contact heat coefficient. It should be noted that, since the area of contact is not known, the coefficient is expressed in terms of unit length (rather than unit area) of contact, so that the dimensions are different from those for the heat-transfer coefficient. Thus, in the case under consideration, the rate of heat flow from metal to graphite per foot length of the fuel elements is 2.5 Δt_{con} Btu/hr, where Δt_{con} is the contact temperature difference between the metal and the graphite in °F.

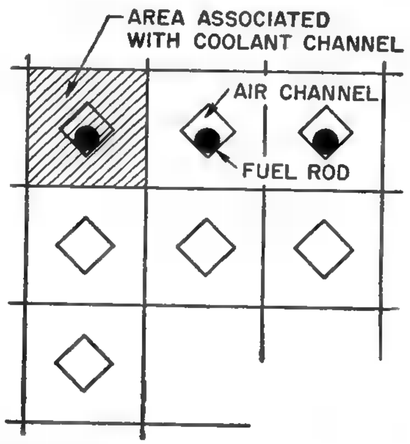


FIG. 11.11 Cylindrical fuel element in square channel in graphite

TEMPERATURE DISTRIBUTION ALONG PATH OF REACTOR COOLANT

GENERALIZED COOLANT CHANNEL

11.102. The foregoing treatment of temperature distribution has dealt exclusively with radial variations; consideration will now be given to the steady-state longitudinal temperature distribution along a fuel element, for example, resulting from the flow of a coolant. The most common type of reactor design involves regularly distributed fuel elements; these are cooled by the flow of some fluid through a number of parallel channels which traverse the entire reactor core. Associated with each channel is an effective area, such as is shown in Fig. 11.11, transverse to the channel length, from which the coolant in the channel removes the heat.

11.103. A generalized representation of a coolant channel, with its associated effective heat-removal area, is shown in Fig. 11.12 in which the different variables are identified. The x-coordinate is seen to represent the direction of coolant flow. The rate, $dq(x)$, at which heat is added to the stream in a differential length, dx, of any coolant passage is then given by

$$dq(x) = wc_p\, dt, \tag{11.103.1}$$

where w is the mass flow-rate (mass/time) of the coolant in the channel, c_p is the specific heat, and dt is the differential temperature increase in the coolant across the length dx. For simplicity it will be postulated that all heat flow within the

solid is normal to the coolant stream, i.e., there is no heat conduction within the solid parallel to the coolant passage.* Then $dq(x)$ is the rate of heat generation in a volume $A\,dx$, where A is the fuel area in the yz-plane associated with the given coolant channel. Since the volumetric heat source is, in general, a point function, $Q^0(x,y,z)$, it is possible to write

$$dq(x) = \left[\int Q^0(x,y,z)\, dy\, dz\right] dx,$$

the integration being carried over the fuel area.

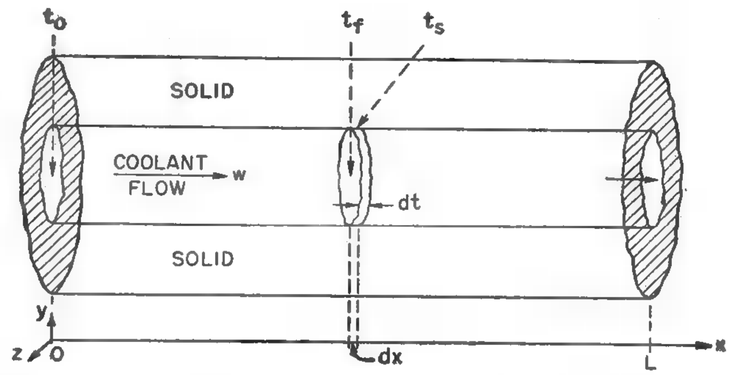


FIG. 11.12 Generalized coolant-flow channel

11.104. If a local average heat source per unit volume, $Q(x)$, is defined by

$$Q(x) = \frac{1}{A}\int Q^0(x, y, z)\, dy\, dz,$$

it follows that

$$dq(x) = AQ(x)\, dx. \tag{11.104.1}$$

Hence, from equations (11.103.1) and (11.104.1), the longitudinal temperature distribution, in the direction of coolant flow, must satisfy the relation

$$\frac{dt}{dx} = \frac{AQ}{wc_p},$$

it being understood that Q is really $Q(x)$. If the quantity A/wc_p is independent of x, the longitudinal distribution of the coolant fluid mixed-mean temperature, t_f, will be given by

$$t_f - t_0 = \frac{A}{wc_p}\int_0^x Q\, dx', \tag{11.104.2}$$

where the fluid entrance temperature, t_0 at $x = 0$, is chosen as the datum. This expression gives the increase in the coolant temperature due to the heat added as it flows along the channel.

* This postulate implies that the following analysis gives a good approximation, provided $dt/dx \ll dt/dn$, where n is the coordinate normal to x.

11.105. By equation (11.67.1), the local (solid) surface temperature, t_s, is related to the mixed-mean fluid temperature, t_f, by

$$\frac{dq}{dA_h} = h(t_s - t_f), \tag{11.105.1}$$

where h is the heat-transfer coefficient for the given conditions, and dA_h is a small element of heat-transfer area. It will be assumed that h is independent of x, which is the case for channels having a large ratio of length to diameter. If p is the passage perimeter, then $dA_h = p\,dx$, and equation (11.105.1) may be written as

$$dq = hp(t_s - t_f)\,dx. \tag{11.105.2}$$

From equations (11.104.1) and (11.105.2) it is seen that

$$t_s - t_f = \frac{AQ}{hp}, \tag{11.105.3}$$

which is the local temperature difference across the solid-coolant convective resistance. If numerator and denominator are multiplied by L, the total channel length, it is found that

$$t_s - t_f = \frac{QV}{hA_h}, \tag{11.105.4}$$

where A_h and V are the heat-transfer area and heat-generating volume, respectively, associated with the channel.

Cosine Longitudinal Source Distribution

11.106. In a bare cylinder or rectangular parallelepiped reactor, the neutron flux has a cosine distribution in the longitudinal direction (Table 3.7). The volumetric heat-source distribution in this direction will thus also be represented by a cosine function (§ 11.39). Previously the origin of the coordinates was chosen at the center of the reactor. For the present purpose, however, it is more convenient to take the origin at the reactor face (Fig. 11.13), so that the heat-source distribution along a particular fuel channel can be represented by

$$Q = Q_0 \sin\frac{\pi x}{L}, \tag{11.106.1}$$

where Q is the volumetric heat-release rate at the point x, and Q_0 is the maximum value at the center of the channel; L is the total channel length, assuming no reflector. Upon inserting this expression for Q into equation (11.104.2) and performing the integration, the result is

$$\begin{aligned} t_f - t_0 &= \frac{Q_0 AL}{\pi w c_p}\left(1 - \cos\frac{\pi x}{L}\right) \\ &= \frac{Q_0 V}{\pi w c_p}\left(1 - \cos\frac{\pi x}{L}\right), \end{aligned} \tag{11.106.2}$$

which gives the rise in temperature of the coolant as it flows through the channel.

11.107. The temperature of the coolant increases continuously, as shown in the bottom curve in Fig. 11.14; the over-all (mixed-mean) temperature rise, represented by Δt_c, is seen to be

$$\Delta t_c = (t_f - t_0)_{x=L} = \frac{2Q_0 V}{\pi w c_p}. \tag{11.107.1}$$

It is of interest to note that this result can be derived in another manner. Since Q_0 is the maximum value of the heat-release rate per unit volume, the average is $2Q_0/\pi$ (cf. § 11.40), and the total heat-release rate for the given channel is then $2Q_0V/\pi$. In the steady state this must, of course, be equal to the rate at which heat is removed by the coolant, i.e., $wc_p\Delta t_c$, so that equation (11.107.1) follows immediately.

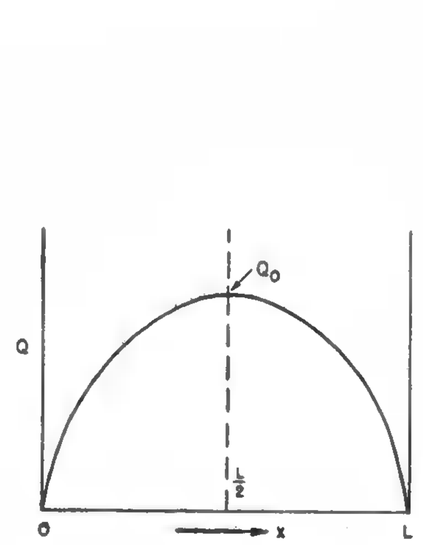


FIG. 11.13 Cosine distribution of volumetric heat source

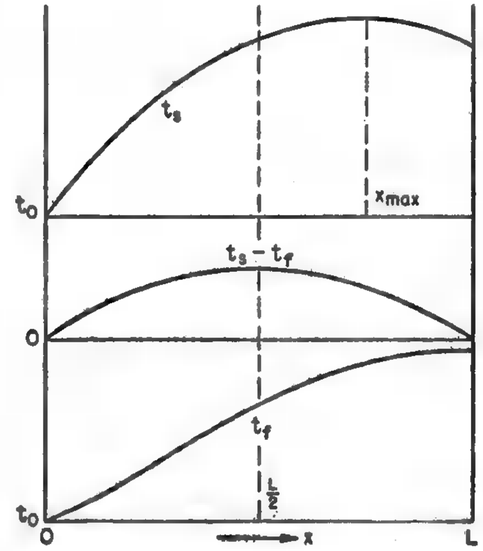


FIG. 11.14 Longitudinal variations of coolant and fuel-surface temperatures

11.108. The temperature difference between the solid and the fluid at any point is obtained from equations (11.105.4) and (11.106.1) as

$$t_s - t_f = \frac{Q_0 V}{hA_h}\sin\frac{\pi x}{L}, \tag{11.108.1}$$

which is seen to be analogous to equation (11.106.1) for the source distribution. The value of $t_s - t_f$, consequently, passes through a maximum when $x = \frac{1}{2}L$, i.e., in the middle of the channel, as shown in the second curve of Fig. 11.14. This result is, of course, to be expected since at any point the fluid-solid temperature difference will be proportional to the radial heat-flow rate at that point, and this is a maximum in the center of the fuel channel.

11.109. The value of $t_s - t_0$, i.e., the metal surface temperature at any point with respect to the value at the channel entrance, is obtained by adding equations (11.106.2) and (11.108.1), so that

$$t_s - t_0 = \frac{Q_0 V}{hA_h} \sin \frac{\pi x}{L} + \frac{Q_0 V}{\pi w c_p}\left(1 - \cos \frac{\pi x}{L}\right). \tag{11.109.1}$$

It is apparent that $t_s - t_0$ must pass through a maximum at some point beyond the middle of the channel, as shown in the top curve of Fig. 11.14. This maximum represents the highest surface temperature of the fuel element. The point at which the maximum is attained is found by differentiating equation (11.109.1) and setting the result equal to zero; thus,

$$x_{\max} = \frac{L}{\pi} \tan^{-1}\left(\frac{-\pi w c_p}{hA_h}\right). \tag{11.109.2}$$

From this expression it is seen that the position of the maximum value of t_s approaches the end of the channel, i.e., $x_{\max} \to L$, as w and c_p decrease, and h and A_h increase.

11.110. The maximum surface temperature in the given channel can now be obtained by insertion of $x_{\max}$ into equation (11.109.1). Assuming the coolant flow rate to be the same in all channels, the maximum fuel element surface temperature occurs in the channel where the neutron flux is a maximum. In this case Q_0 is the volumetric heat-release rate at the center of the reactor, and the corresponding maximum value of t_s may determine the maximum reactor power.

11.111. It should be noted that the surface temperature, t_s, considered above, is less than the temperature in the middle of the fuel element. The difference, represented by the temperature drop through the fuel region and the cladding, can be calculated by the methods described earlier (cf. § 11.81, *et seq.*).

11.112. For purposes of computation, it is convenient to represent $\pi x/L$ by the symbol α, i.e.,

$$\alpha \equiv \frac{\pi x}{L}.$$

It can then be found from equation (11.109.2) that

$$\tan \alpha_{\max} = -\frac{\pi w c_p}{hA_h}, \tag{11.112.1}$$

where $\alpha_{\max} = \pi x_{\max}/L$. Utilizing this expression for $\tan \alpha_{\max}$, equation (11.109.1) can be reduced to

$$(t_s)_{\max} - t_0 = \frac{Q_0 V}{\pi w c_p}(1 - \sec \alpha_{\max}). \tag{11.112.2}$$

With these two equations the value of the maximum surface temperature and its location can be readily evaluated.

Example Flat fuel elements, 2.5 in. wide and 30 in. long, are arranged in an approximately cylindrical array in a water-cooled (and moderated) reactor, of the Swimming

Pool type. To prevent the water from boiling, the maximum surface temperature of the elements is set at 200°F. The water enters at 100°F and flows upward at a rate of 3 ft/sec through cooling channels formed by spacing the fuel plates at 0.120 in. apart (Fig. 11.15). Assuming for simplicity that the water reflector has a negligible effect on the neutron flux in the reactor, determine (*a*) the minimum number of channels required if the reactor is to operate at 3000 kw, and (*b*) the average temperature of the exit water under these conditions. Given that at the metal-water interface $h = 1050$ Btu/(hr)(ft²)(°F); the density of the water is 61.8 lb/ft³, and the specific heat is 1.00 Btu/(lb)(°F). (The heating of the water in its capacity as moderator may be neglected.)

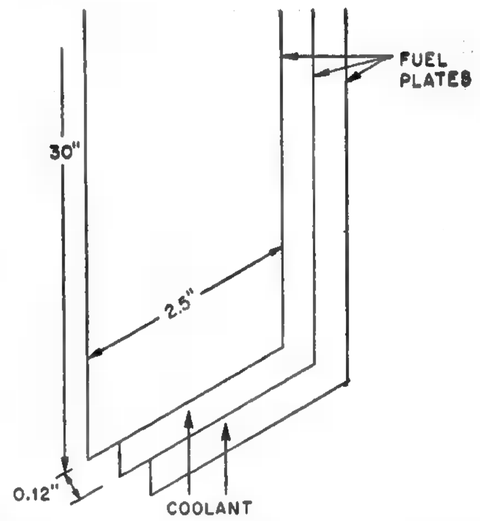


FIG. 11.15 Coolant channels between vertical fuel plates

The mass rate of flow, w, of the coolant per channel is equal to $v\rho A_f$, where v is the velocity of the coolant (3 ft/sec = 10,800 ft/hr), ρ is the density of water (61.8 lb/ft³), and A_f is the cross-sectional (or flow) area of a coolant channel. In this case $A_f = (2.5)(0.12)/144 = 2.08 \times 10^{-3}$ ft²; hence,

$$w = v\rho A_f$$
$$= (10{,}800)(61.8)(2.08 \times 10^{-3}) = 1.39 \times 10^3 \text{ lb/hr}.$$

In order to determine $x_{\max}$, the value of A_h, the heat-transfer area per channel, is required. Since the coolant in each channel receives heat from two fuel-element surfaces, it follows that

$$A_h = \frac{(2)(2.5)(30)}{144} = 1.04 \text{ ft}^2.$$

Consequently, by equation (11.112.1),

$$\tan \alpha_{\max} = -\frac{(3.14)(1.39 \times 10^3)(1.00)}{(1050)(1.04)} = -3.98$$

and, hence,

$$\sec \alpha_{\max} = -4.11.$$

The problem stipulates that $(t_s)_{\max}$ is 200°F and, since t_0 is given as 100°F, it follows from equation (11.112.2) that

$$Q_0 V = 100 \times \frac{(3.14)(1.39 \times 10^3)(1.00)}{5.11} = 8.55 \times 10^4 \text{ Btu/(hr)(channel)}.$$

The value of Q_0, the volumetric heat source at the center of the core, may be taken to be 3.64 times the average; hence,

$$Q_{av} V = \frac{8.55 \times 10^4}{3.64} = 2.35 \times 10^4 \text{ Btu/(hr)(channel)},$$

which gives the average rate of heat generation per channel. The total power of the reactor is to be 3000 kw, i.e., 10.24×10^6 Btu/hr, and so the minimum number, n, of coolant channels required is

$$n = \frac{10.24 \times 10^5}{2.35 \times 10^4} = 435 \text{ channels.}$$

The average temperature rise of the coolant, Δt_c, is obtained by equating $Q_{av}V$ (heat generated per channel) to $wc_p\Delta t_c$ (heat taken up by coolant per channel); then

$$\Delta t_c = \frac{Q_{av}V}{wc_p} = \frac{2.35 \times 10^4}{(1.39 \times 10^3)(1.00)} = 16.9°\text{F}.$$

11.113. Although the foregoing treatment is adequate for preliminary calculations, its limitations should be borne in mind. In the first place, the results apply to a bare core without reflector and so are not truly applicable to a Swimming Pool type reactor. For a reflected reactor such as this, the flux (or power) distribution is flatter than in an equivalent bare core, and the ratio of maximum to average specific power (or volumetric heat source) is lower. One method of treating the problem of a reactor with a thick reflector is to assume a cosine source distribution, as in Fig. 11.13, but to suppose that the value of Q goes to zero at some distance, e.g., about one or two reflector diffusion lengths, beyond the boundary of the core. The integration of $\sin \pi x/L$, referred to in § 11.106, then does not start from zero, but from x equal to roughly two diffusion lengths in the reflector.

11.114. In addition, there are other factors, of a physical nature, which affect the accuracy of the calculations. For example, the coolant flow rate may not be the same in all channels, and the over-all heat-transfer coefficient may vary, due to such causes as corrosion of surfaces, deposits of scale, etc. Further, conduction along the axis of the fuel elements and the variation of h along the channel, which were neglected, may have an equally important effect on the results.

HEAT TRANSFER CHARACTERISTICS OF FLUIDS

Laminar and Turbulent Flow

11.115. The study of the flow of fluids, in connection with reactor systems, is important for several reasons. As seen in earlier sections, all the heat generated in a reactor is removed and transferred from one medium to another either as sensible heat or (in a boiling reactor) as latent heat. These processes clearly involve the flow of one or more fluids. From the properties of these fluids and their flow characteristics, it is usually possible to make reasonable predictions concerning the heat-transfer coefficients for simple geometries (§ 11.124). For complex cases, however, the experimental determination of the heat-transfer coefficients is required.

11.116. In addition, a knowledge of the pressure drops in various parts of the cooling system is necessary in order to establish the over-all pumping power requirements. The methods whereby the pressure drops are estimated in a number of cases will be described below.

11.117. Before proceeding with the more detailed aspects of the problems of interest, the essential nature of the two important types of fluid flow will be reviewed briefly. When a fluid flows through a straight pipe at low velocity, the particles of fluid move in straight lines parallel to the axis of the pipe, without any appreciable radial motion. This is described as laminar, streamline, or viscous flow, and the "velocity profile" is depicted diagrammatically in Fig. 11.16, A; the lengths of the arrows indicate the fluid velocity at various points across the pipe. The velocity distribution curve in a circular pipe is a parabola, and the average velocity is half the maximum at the center of the pipe.

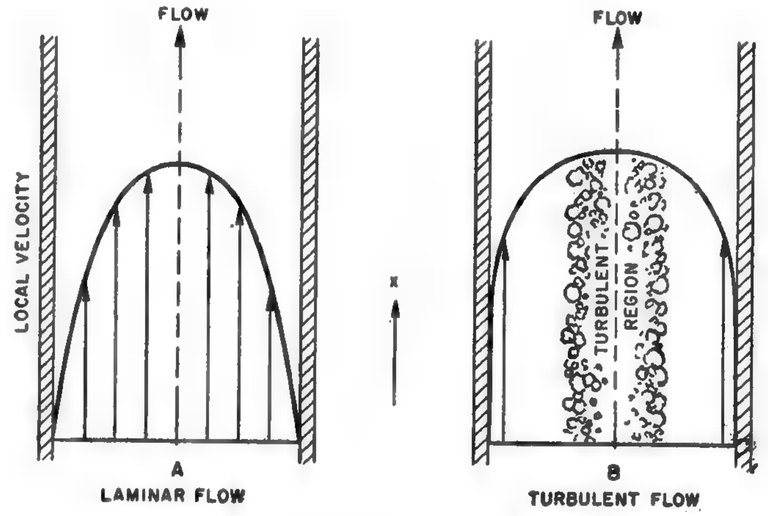


Fig. 11.16 A. Laminar flow
B. Turbulent flow

11.118. One characteristic of streamline flow is that it obeys Newton's equation*

$$F = \mu A \frac{du}{dy}, \tag{11.118.1}$$

where F is the shearing force (or fluid friction) over an area A between two parallel layers of fluid, moving with different velocities in a region where the velocity gradient is du/dy, perpendicular to the direction of flow; the symbol μ represents the absolute (or dynamic) viscosity of the fluid, which is defined by equation (11.118.1). The dimensions of viscosity are seen to be mass/(length)(time); in the cgs system, the unit of viscosity, expressed in g/(cm)(sec), is called the poise, and it is in terms of this unit (or its hundredth part, called a centipoise) that viscosity values are frequently calculated. In the English (or engineering) system of units, the dynamic viscosity is conveniently expressed

* Certain fluids, e.g., suspensions, which do not obey the Newton equation, are referred to as non-Newtonian fluids.

as lb/(ft)(hr); the values in this system are obtained upon multiplying the number of centipoises by 2.42.

11.119. Laminar flow may be distinguished from other types of flow by means of a dimensionless quantity called the Reynolds number (or modulus); this is represented by Re and defined by

$$\mathrm{Re} \equiv \frac{Dv\rho}{\mu}, \tag{11.119.1}$$

where D is the pipe diameter, v is the mean velocity of the fluid, ρ is its density, and μ its viscosity. Experiments with many fluids have shown that, in general, flow is laminar, and Newton's equation is obeyed, in ducts of uniform cross section as long as Re is less than about 2100. The precise critical value of the Reynolds number depends, to some extent, on the flow conditions.

11.120. If the circumstances are such that Re exceeds about 4000 in such systems, the fluid motion is turbulent; this type of flow is characterized by the presence of numerous eddies which cause a radial motion of fluid, i.e., motion across the stream, in addition to the flow parallel to the pipe axis. Newton's equation is then no longer valid.

11.121. The velocity profile for turbulent flow is shown in Fig. 11.16, B. It appears that the three more-or-less distinct regions exist in a turbulent stream. First, a layer near the wall in which the flow is essentially laminar, as indicated by the arrows. Then a transition (or buffer) zone in which some turbulence exists, and finally there is the fully turbulent core. The local velocity of flow increases toward the pipe axis, at first more rapidly and then more slowly than for completely laminar motion. As a result, there is a marked flattening of the velocity profile; this flattening becomes more pronounced with increasing Reynolds number. The ratio of the mean flow velocity to the maximum ranges from about 0.75 to 0.81, as Re increases from 5000 to 100,000.

11.122. The Reynolds number, as given by equation (11.119.1), is applicable only to pipes of circular cross section; for noncircular channels, especially long rectangular ducts and annular spaces, a good approximation is obtained if D in equation (11.119.1) is replaced by the equivalent (or hydraulic) diameter, D_e, defined by

$$D_e = 4 \times \frac{\text{Cross section of stream}}{\text{Wetted perimeter of duct}}, \tag{11.122.1}$$

which becomes identical with the actual diameter for a circular pipe. For the channel between two parallel plates, as in reactors of the Swimming Pool type, D_e is approximately twice the distance between the plates (see following example).

Example Determine the Reynolds number for the cooling water of the reactor in the example in § 11.112.

The channel cross section is (2.5 in.)(0.12 in.), and 2.5 in. on each side plus 0.12 in. on each end is wetted; hence,

$$D_e = 4 \times \frac{(2.5)(0.12)}{(2)(2.5 + 0.12)} = 0.23 \text{ in.}$$

i.e., roughly twice the distance between the plates. The mean flow velocity may be taken as (3)(3600) = 10,800 ft/hr; the density of water is 61.8 lb/ft³; and the viscosity at 100°F is 1.66 lb/(hr)(ft); hence

$$\mathrm{Re} = \frac{(0.02)(10{,}800)(61.8)}{1.66} = 8050.$$

The flow is thus definitely in the turbulent region.

11.123. In the range between the Re values of 2100 and 4000 there is generally a transition from purely laminar flow to complete turbulence. Laminar behavior sometimes persists in this region or turbulence may increase, depending, among other factors, on entrance conditions, the occurrence of upstream turbulence, the roughness of the pipe, the presence of obstacles, and on pulsations in flow rate produced by the pump or some other component in the system.

Prediction of Heat-transfer Coefficient for Turbulent Flow of Ordinary Fluids*

11.124. In essentially all cases of reactor cooling, where forced convection is used, the coolant is under turbulent flow conditions.† Satisfactory predictions of heat-transfer coefficients in long straight channels of uniform cross section can be made on the assumption that the only variables involved are the mean velocity of the fluid coolant, the diameter (or equivalent diameter) of the coolant channel, and the density, heat capacity, viscosity, and thermal conductivity of the coolant. From the fundamental differential equations or by the use of the methods of dimensional analysis,‡ it can be shown that heat-transfer coefficients can be expressed in terms of three dimensionless moduli; one of these is the Reynolds number, already defined, and the others are the Nusselt number (Nu) and the Prandtl number (Pr), defined by

$$\mathrm{Nu} \equiv \frac{hD}{k} \quad \text{and} \quad \mathrm{Pr} \equiv \frac{c_p\mu}{k}.$$

11.125. As a result of numerous experimental studies of heat transfer, various expressions relating these three moduli have been proposed; one of these, for a coolant fluid in a long straight channel, is §

* For treatment of heat transfer in laminar flow, see McAdams, *op. cit.*, p. 229, *et seq.*

† Some reactors, e.g., the CP-2 (uranium-graphite) reactor, the Canadian ZEEP (uranium-heavy water), and the British fast reactor (ZEPHYR), which operate at very low (almost zero) power, and the reactors of the Swimming Pool type at powers of 100 kw or less, make use of conduction and free convection for cooling.

‡ See, for example, the standard texts referred to in ‖ 11.61.

§ F. W. Dittus and L. M. K. Boelter, *Univ. of Calif. Pub. Eng.*, 2, 443 (1930); see also A. P. Colburn, *Trans. Am. Inst. Chem. Engrs.*, 29, 174 (1933); W. H. McAdams, *op. cit.*, p. 219.

$$\frac{hD}{k} = 0.023\left(\frac{Dv\rho}{\mu}\right)^{0.8}\left(\frac{c_p\mu}{k}\right)^{0.4} \qquad (11.125.1)$$

or

$$\mathrm{Nu} = 0.023\ \mathrm{Re}^{0.8}\ \mathrm{Pr}^{0.4}. \qquad (11.125.2)$$

Since, for normal flow rates, the resistance to heat transfer is mainly in the laminar layer of liquid adjacent to the surface, the temperature of this layer is that at which the various physical properties, especially the highly temperature-sensitive viscosity, should be evaluated. For practical purposes this may be taken as the mean of the surface and bulk fluid temperatures.

11.126. It is seen from equation (11.125.1), that, if the viscosity, thermal conductivity, density, and specific heat of the coolant are known, the heat-transfer coefficient can be estimated for turbulent flow of given velocity in a pipe or channel of specified diameter (or equivalent diameter). The results appear to be satisfactory for values of Re in excess of about 10,000, and for Pr values from 0.7 to 120. This range of Prandtl numbers includes gases and essentially all liquids, except liquid metals; the latter have very low Prandtl numbers, primarily because of their high thermal conductivity but also often because of their low viscosity and heat capacity. The correlations for liquid metals will, therefore, be considered separately (§ 11.128). For gases, Pr is approximately 0.75 and so equation (11.125.2) reduces to

$$\mathrm{Nu} \approx 0.020\ \mathrm{Re}^{0.8}\ \text{(for gases)}.$$

11.127. Where large differences exist between the temperatures of the solid and of the fluid, the associated variations in the physical properties of the coolant can influence the heat transfer. As indicated above, such variation is most significant for the viscosity, and in order to make allowances for it the relationship

$$\mathrm{Nu} = 0.027\ \mathrm{Re}^{0.8}\ \mathrm{Pr}^{0.33}\left(\frac{\mu}{\mu_w}\right)^{0.14}$$

has been proposed, where μ is the viscosity at the mixed-mean fluid temperature and μ_w is that at the wall temperature.*

11.128. For water as coolant, allowance for the variations in physical properties with temperature may be made by writing

$$h\ \mathrm{Btu/(hr)(ft^2)(°F)} = 170(1 + 10^{-2}t - 10^{-5}t^2)\frac{v^{0.8}}{D^{0.2}}, \qquad (11.128.1)$$

where, it should be noted, v is in ft/sec and D in in.; the temperature is t°F.† The values for the heat-transfer coefficient given by this relationship are in good agreement with those obtained from equation (11.125.1) for temperatures up to 600°F.

* E. N. Sieder and G. E. Tate, *Ind. Eng. Chem.*, **28**, 1429 (1986).
† J. A. Lane, unpublished.

Example Calculate the heat transfer coefficient for the water coolant in the problem following § 11.112 using (*a*) the Dittus-Boelter equation and (*b*) equation (11.128.1).

The temperature is taken as 100°F so that $\rho = 61.8$ lb/ft³, $c_p = 1.00$ Btu/(lb)(°F), and μ is 1.66 lb/(ft)(hr). From the table in the Appendix, k for water is 0.363 Btu/(hr)(ft²)(°F/ft). The value of v is 10,800 ft/hr and the (equivalent) diameter of the duct is 0.02 ft. Hence, from the Dittus-Boelter equation,

$$h = 0.023 \times \frac{0.363}{0.02}\left[\frac{(0.02)(10{,}800)(61.8)}{1.66}\right]^{0.8}\left[\frac{(1.00)(1.66)}{0.363}\right]^{0.4}$$
$$= 1020\ \mathrm{Btu/(hr)(ft^2)(°F)}.$$

By equation (11.128.1) for water

$$h = (170)[1 + (10^{-2})(100) - (10^{-5})(100)^2]\frac{(3)^{0.8}}{(0.24)^{0.2}}$$
$$= (170)(1.9)\frac{2.41}{0.752}$$
$$= 1035\ \mathrm{Btu/(hr)(ft^2)(°F)}.$$

Liquid-metal Heat-transfer Coefficient for Turbulent Flow*

11.129. Because of their high thermal conductivity, mentioned earlier, the Prandtl numbers of liquid metals may be as low as 0.005. The controlling resistance to the radial transfer of heat is not across the laminar sub-layer, as it is for other liquids, but it may extend into a major part of the turbulent core. It is to be expected, therefore, that relationships applicable to gases and non-metallic liquids will not apply to liquid metals. For the correlation of heat-transfer data it is now appropriate to use another dimensionless modulus, namely, the Peclet number, Pe, which is the product of the Reynolds and Prandtl numbers; thus,

$$\mathrm{Pe} \equiv \mathrm{Re} \times \mathrm{Pr} \equiv \frac{Dv\rho c_p}{k}.$$

For the heat transfer to liquid metals flowing turbulently in circular pipes, with uniform heat flux, the relationship

$$\frac{hD}{k} = 7 + 0.025\left(\frac{Dv\rho c_p}{k}\right)^{0.8} \qquad (11.129.1)$$

or

$$\mathrm{Nu} = 7 + 0.025\ \mathrm{Pe}^{0.8} \qquad (11.129.2)$$

has been suggested for the estimation of heat-transfer coefficients.† It will be observed that, according to this equation, the heat transfer is independent of the viscosity of the liquid metal coolant. This is so because in such a fluid thermal conduction is more important than mass motion in transferring heat.

* See R. N. Lyon (Ed.), "Liquid-Metals Handbook," 2nd ed., U. S. Government Printing Office, 1952, Chapter 5 (by R. N. Lyon and H. F. Poppendiek).
† R. N. Lyon, *Chem. Eng. Prog.*, **47**, 75 (1951).

11.130. Equation (11.129.1) appears to predict h reasonably well for alkali metals at moderately high temperatures, but for other liquid metals, e.g., lead, bismuth, and mercury, the calculated values are frequently higher than those obtained by experiment, possibly due to the nonwetting characteristics of these metals. For this reason it is recommended that the heat-transfer coefficient used in designing equipment for the heavier and less chemically active metals be taken as half the value obtained from equation (11.129.1).*

11.131. When conduction is the predominant heat-transfer mechanism, as is the case for liquid metals, the equivalent-diameter concept, referred to in § 11.122, is not valid. A modified expression, which is effectively the result of multiplying the right side of equation (11.129.2) by 0.8, namely,

$$\mathrm{Nu} = 5.8 + 0.020\ \mathrm{Pe}^{0.8},$$

has been proposed for flow in the channel between parallel plates with heat being removed uniformly from one side only.† The equivalent diameter used in computing Nu and Pe is taken as twice the distance between the plates (§ 11.122).

11.132. For liquid metals flowing in annular channels, with a uniform wall heat flux, a good approximation appears to be

$$\mathrm{Nu} = 5.3 + 0.019\ \mathrm{Pe}^{0.8}\left(\frac{r_o}{r_i}\right)^{0.3},$$

where r_o and r_i are the outer and inner radii, respectively, of the annulus.‡ It should be noted that, when using the two foregoing equations for design calculations, it is advisable to apply the same safety factor, i.e., 0.5, as for equation (11.129.1) when dealing with the heavy metals.

11.133. An interesting fact, to which attention may be called here, is that, although the heat-transfer coefficients for both nonmetals and metals are increased by increasing the rate of flow of the coolant, the effect is relatively small for liquid metals. An examination of equation (11.125.1) shows that for a nonmetal h is proportional to $v^{0.8}$, but from equation (11.129.1) for a liquid metal it is seen that the term containing $v^{0.8}$ is added to a fairly large term which is independent of the flow velocity. The physical basis of this behavior is that, even under turbulent flow conditions, a large proportion of the heat transferred from a solid surface to a liquid metal is by conduction and this is not affected by the flow rate of the coolant.

Comparison of Heat-transfer Characteristics of Coolants §

11.134. A discussion of reactor coolants, mainly from the standpoints of their neutron capture cross sections and radiation stability, was given in Chapter

* R. N. Lyon, unpublished.
† R. A. Seban, *Trans. Am. Soc. Mech. Engrs.*, **72**, 789 (1950).
‡ R. V. Bailey, Report ORNL-521.
§ For literature references, see § 11.168.

VIII. It is appropriate here to examine their thermal properties. A good coolant is, in general, one having a high heat-transfer coefficient (and a large volumetric heat capacity) under the conditions that exist (or can exist) in a reactor. Thus, if h is large, it is possible to realize a specified rate of heat removal from the fuel elements with a relatively small heat-transfer area. It is also possible to decrease the rate of flow of the coolant and, as a result, the power required for pumping (§ 11.162). The decrease in the flow rate will decrease the heat-transfer coefficient, but if the latter is sufficiently high, in any case, the decrease may not be serious, especially for a liquid metal.

11.135. It can be seen from equations (11.125.1) and (11.129.1) that, for a given flow rate and coolant channel diameter, large heat-transfer coefficients are associated with high thermal conductivity, density, and heat capacity. In addition, for nonmetallic fluids, a low viscosity is advantageous. It is apparent, from general considerations, therefore, that water and many liquid metals may be expected to have good heat-transfer characteristics. Such materials will therefore permit considerable flexibility in the design of the heat-removal system of the reactor.

11.136. Gases have the disadvantage of low density and thermal conductivity, but some compensation is achieved, as far as the former property is concerned, by operating at high pressures. The high flow rate required with a gaseous coolant (§ 11.164) also helps to increase the heat-transfer coefficient although at the cost of very significant increase in pumping power.

11.137. Another basis for the comparison of coolants is what is called the volumetric heat capacity. The total heat release rate, q_{tot}, of the reactor is related to the total mass flow rate of the coolant, w, its specific heat, c_p, and the average rise, Δt_c, in the mixed-mean temperature of the coolant by

$$q_{\mathrm{tot}} = wc_p\Delta t_c.$$

Since w may be replaced by $v\rho A_f$, where A_f is here the total flow area, it follows that

$$q_{\mathrm{tot}} = (\rho c_p)vA_f\Delta t_c. \tag{11.137.1}$$

The quantity ρc_p is called the *volumetric heat capacity*, which can be expressed in Btu/(ft³)(°F). For a given coolant flow rate, v, and total flow area, A_f, a high volumetric heat capacity will make possible a specified rate of heat removal without an excessive (average) rise in the coolant temperature. This is advantageous from the thermal stress standpoint. Although liquid metals may be somewhat superior to water with respect to heat transfer, the latter has a better volumetric heat (or heat-transport) capacity. Thus, at 400°F, ρc_p for water is 53.7, whereas that for liquid sodium is 17.1 Btu/(ft³)(°F). At 10-atm pressure the volumetric heat capacity of carbon dioxide gas is only 0.174 Btu/(ft³)(°F), so that very high flow rates are necessary with gaseous coolants.

Example Compare the heat-transfer coefficient for turbulent flow at 400°F of (*a*) carbon dioxide at 10 atm and a velocity of 100 ft/sec, (*b*) water at 20 ft/sec, and (*c*) liquid sodium at 20 ft/sec, flowing through a long, straight, circular tube of diameter 0.10 ft.

The physical properties of the three coolants at 400°F are tabulated below:

	Carbon Dioxide	*Water*	*Liquid Sodium*
ρ, lb/ft³	0.656*	53.7	53.2
μ, lb/(hr)(ft)	0.0540	0.148	—
k, Btu/(hr)(ft²)(°F/ft)	0.0184	0.380	47.1
c_p, Btu/(lb)(°F)	0.265	1.00	0.320

For *carbon dioxide*, using equation (11.125.1)

$$h = \frac{0.0184}{0.10} \times 0.023 \left[\frac{(0.1)(100 \times 3600)(0.656)}{0.0540}\right]^{0.8} \left[\frac{(0.265)(0.0540)}{0.0184}\right]^{0.4}$$
$$= 124 \text{ Btu/(hr)(ft²)(°F)}.$$

For *water*, from the same equation (11.125.1)

$$h = \frac{0.380}{0.10} \times 0.023 \left[\frac{(0.1)(20 \times 3600)(53.7)}{0.148}\right]^{0.8} \left[\frac{(1.00)(0.148)}{0.380}\right]^{0.4}$$
$$= 8150 \text{ Btu/(hr)(ft²)(°F)}.$$

For *liquid sodium*, however, equation (11.129.1) is used; thus,

$$h = \frac{47.1}{0.10}\left\{7 + 0.025\left[\frac{(0.10)(20 \times 3600)(53.2)(0.320)}{47.1}\right]^{0.8}\right\}$$
$$= 9650 \text{ Btu/(hr)(ft²)(°F)}.$$

Heat Transfer from Fluids Containing a Volume Heat Source

11.138. Because reactors with circulating fuel have certain advantages (§ 12.132), they are attracting interest for possible use in power production. A consideration of heat transfer in fluid fuels is, therefore, an important aspect of reactor design. Since the fluids (while in the reactor) contain heat sources due to fission, the determination of the temperature distribution in the flowing system and at the walls involves unusual principles of heat transfer and fluid flow. These principles will be reviewed briefly, as a complete treatment would require a much more detailed knowledge of heat transfer and the theory of fluid flow than has been presented in this chapter.†

11.139. Consider a reactor with a circulating fluid fuel in which heat is being generated. A possible example would be the Liquid Metal Fuel Reactor (LMFR), in which a solution of uranium in liquid bismuth is the fuel circulated through channels in the graphite moderator.‡ If the fuel is forced through a channel or pipe in the reactor, the heat will be convected away relatively slowly by the slow-moving part of the stream close to the wall. As a result, there

* Calculated from the ideal gas laws.

† See H. F. Poppendiek, *Chem. Eng. Prog. Symposium Series*, No. 11, **50**, 93 (1954).

‡ For reference, see § 8.246.

will be a temperature rise toward the wall of the channel, and a flow of heat from the slower-moving parts of the stream to the faster-moving parts near the middle of the stream. If the temperature of the wall is limited, some prediction of the difference between the wall temperature and the mixed-mean fluid temperature is required.

11.140. Mathematical expressions have been derived for determining this temperature for an idealized system which is defined, essentially, by the following postulates: (1) thermal and hydrodynamic boundary layers are established, i.e., the flow is in long pipes; (2) the volume heat source within the fluid is uniform; (3) heat is transferred uniformly from (or to) the liquid at the pipe wall; (4) physical properties are independent of temperature; and (5) in the case of turbulent flow, the hydrodynamic structure is defined by a generalized velocity profile.

11.141. The differential equation describing heat transfer in a pipe for the general case of turbulent flow is

$$u(r)r\frac{dt}{dx} = \frac{\partial}{\partial r}\left\{\left[\alpha + \epsilon(r,u)\right] r\frac{\partial t}{\partial r}\right\} + \frac{Qr}{\rho c_p}, \qquad (11.141.1)$$

where r is the radial distance from the pipe center line and x is the axial distance, $u(r)$ is the fluid velocity, α is the fluid thermal diffusivity, and ϵ is the eddy diffusivity;* Q is the rate of heat release per unit volume, as before, and t, c_p, and ρ have their usual significance. The two boundary conditions for the problem are (1) the uniform wall heat flux is given and (2) some system reference temperature, e.g., the mixed-mean fluid temperature or the center-line temperature, is known.

11.142. Upon substituting into equation (11.141.1) an expression for the axial temperature gradient in the region of established flow, a total differential equation results. This boundary value problem can be separated into two somewhat simpler problems whose solutions can be superposed to yield the solution to the original problem. One of these simpler problems pertains to turbulent flow in a pipe with a uniform volumetric heat source within the fluid but with zero heat flux at the wall, whereas the second applies to turbulent flow in a pipe without an internal heat source, but with a uniform wall heat flux. Solutions of these problems for the temperature distribution have been obtained in terms of the dimensionless moduli considered above.†

11.143. In superposing the solutions to these two problems, temperature differences, rather than actual temperatures are added. For example, the calculated values of the difference between the temperature of the fluid at the pipe wall (t_w) and the mixed-mean fluid temperature (t_f) at any point, for the two

* Cf., McAdams, *op. cit.*, pp. 34, 209. In the above treatment, it is postulated that the heat- and momentum-transfer eddy diffusivities are equal.

† The solution to the first boundary problem is given by H. F. Poppendiek, *loc. cit.*; several solutions to the second problem are available, e.g., R. C. Martinelli, *Trans. Am. Soc. Mech. Engrs.*, **69**, 947 (1947).

separate problems, are added to give the corresponding difference for the actual system involving turbulent flow in a pipe with a uniform volumetric heat source and a uniform wall heat flux.

11.144. The results obtained for the case of turbulent flow of a fluid containing a uniform heat source, but with no cooling at the pipe wall, is depicted in Fig. 11.17. Here the dimensionless "temperature difference," $(t_w - t_f)/(Qr_0^2/k)$, where r_0 is the pipe radius, is shown as a function of the Reynolds number for certain specified values of the Prandtl modulus. Experimental measurements

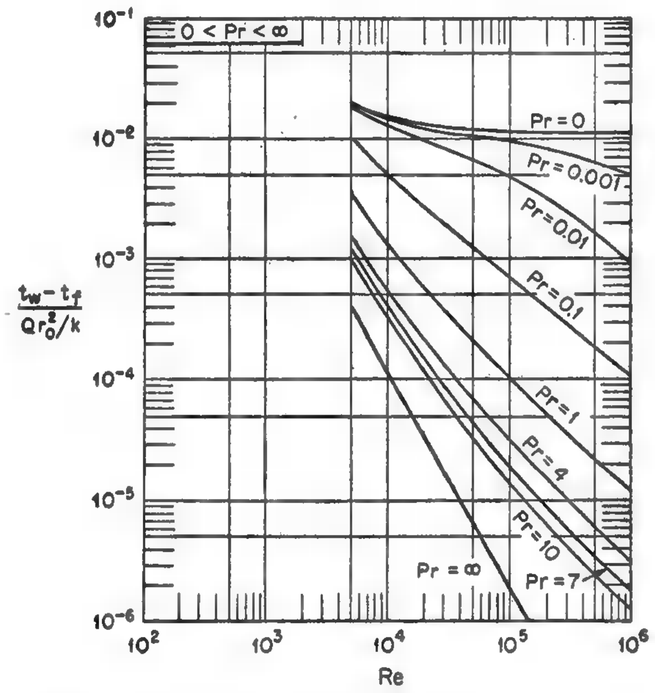


FIG. 11.17 Dimensionless wall-fluid temperature difference as function of Reynolds and Prandtl numbers (H. F. Poppendiek)

have confirmed the reliability of the calculations in the range of Pr = 5 to 10 and Re = 750 to 14,000. The use of Fig. 11.17 is illustrated by the following example.

Example A liquid fuel is circulated through pipes of circular cross section under the following (turbulent flow) conditions: Re = 10,000; Pr = 10; $Q = 9.66 \times 10^7$ Btu/(hr)(ft³); $k = 1.5$ Btu/(hr)(ft²)(°F/ft); and $r_0 = 0.166$ ft. Determine (*a*) the difference between the wall temperature and the mixed-mean fluid temperature for an uncooled wall; (*b*) the wall cooling required to reduce the wall temperature to the mixed-mean fluid temperature; and (*c*) the fraction of the heat generated in the bulk of the fluid that would have to be extracted to achieve this cooling.

(*a*) From Fig. 11.17, for the case of no heat removal at the wall, it is seen that when Re = 10,000 and Pr = 10,

$$\frac{t_w - t_f}{Qr_0^2/k} = 3.1 \times 10^{-4}.$$

Hence, the required temperature difference is given by

$$t_w - t_f = 3.1 \times 10^{-4} \frac{Qr_0^2}{k}$$

$$= \frac{(3.1 \times 10^{-4})(9.66 \times 10^7)(0.166)^2}{1.5}$$

$$= 550°\text{F}.$$

(*b*) The heat-transfer coefficient for this pipe system is obtained from equation (11.125.2), noting that D is equal to $2r_0$; hence,

$$h = \left(\frac{k}{2r_0}\right) 0.023 \text{ Re}^{0.8} \text{ Pr}^{0.4}$$

$$= \frac{1.5}{(2)(0.166)} (0.023)(10{,}000)^{0.8}(10)^{0.4}$$

$$= 413 \text{ Btu/(hr)(ft}^2\text{)(°F)}.$$

The heat flux at the wall, represented by $(q/A)_0$, required to make the wall temperature equal to the mixed-mean fluid temperature, is obtained by equating this flux to $h\Delta t$, where Δt is the value of $t_w - t_f$ for no heat flux at the wall, i.e., 550°F. It is seen, therefore, that

$$\left(\frac{q}{A}\right)_0 = h\Delta t = (413)(550)$$

$$= 227{,}000 \text{ Btu/(hr)(ft}^2\text{)}.$$

(*c*) For unit length of pipe, the heat extracted at the wall is equal to $2\pi r_0(q/A)_0$ Btu/hr, whereas the heat generated in the fluid volume is $\pi r_0^2 Q$ Btu/hr. The fraction of the heat generated that would be extracted by wall cooling is then

$$\frac{2\pi r_0(q/A)_0}{\pi r_0^2 Q} = \frac{(2)(3.14)(0.166)(2.27 \times 10^5)}{(3.14)(0.166)^2(9.66 \times 10^7)}$$

$$= 0.0282,$$

which is 2.8 per cent.

(It is of interest to note that had the flow in the pipes been laminar, the value of $t_w - t_f$ without cooling could have been about 200 times as great as for turbulent flow. The corresponding rate of cooling at the wall would have to be roughly ten times as large.)

HEAT TRANSFER FROM SLURRIES

11.145. Some reference may be made to heat transfer from slurries (or suspensions) since they have been proposed as the circulating fuel and blanket media for homogeneous reactors with external cooling. If the slurries are Newtonian fluids, they can be treated as ordinary fluids, provided, of course, precautions are taken to prevent settling or caking-out of the solid matter on the conduit walls.

11.146. If the slurries are non-Newtonian in their behavior, they will usually be either plastic or pseudo-plastic, in which case the effective viscosity will

depend on the shear rate at each point. It appears from pressure-drop and heat-transfer measurements that, under turbulent flow conditions, a given plastic or pseudo-plastic slurry frequently has essentially a constant effective viscosity which is, in some instances, approximately equal to the viscosity of the liquid vehicle. Substitution of such an effective viscosity for the fluid viscosity in the equations for ordinary fluids, e.g., equation (11.125.1), then usually gives reasonably good values for the heat-transfer coefficient.

11.147. In general, however, if slurries are to be used, it is probably necessary that sufficient experiments be made to check the predicted heat-transfer coefficients. It is also necessary to determine whether caking does (or does not) occur on the heat-transfer surface. In addition, particular care should be taken in connection with passages containing constricted regions, since a true plastic material may become stationary in such regions.

Heat-transfer Coefficients in Free Convection

11.148. The prediction of heat-transfer coefficients associated with free (or natural) convection is much more difficult than for forced convection. In some systems, however, e.g., Swimming Pool reactors at low power, free convection is important for reactor cooling.* In these cases there is an upward motion of the coolant due to buoyancy resulting from heat transferred to the fluid by conduction.

11.149. For free convection a further dimensionless modulus, the Grashof number, Gr, is used; this is defined by

$$\mathrm{Gr} \equiv \frac{L^3\rho^2 g\beta\Delta t}{\mu^2},$$

where L is an appropriate length (or diameter), g is the acceleration of gravity, β is the thermal coefficient of volume expansion of the fluid, and Δt is a prescribed temperature difference within the fluid; ρ and μ are, as before, the density and viscosity of the fluid. For laminar free convection from either a vertical plate or a large cylinder whose surface is at a uniform temperature and which is in an infinite body of fluid, the local Nusselt number is related to the Grashof and Prandtl moduli by

$$\mathrm{Nu}_x = \frac{hx}{k} = 0.55(\mathrm{Gr}_x\,\mathrm{Pr})^{0.25}, \qquad (11.149.1)$$

where the subscript x indicates that the heat-transfer coefficient is evaluated at a point on the plate at a distance x from the bottom. In evaluating Nu_x and Gr_x, this distance x is used for the length dimension, i.e., for D or L. The average value of Nu is obtained by integration over the total height of the plate or cylinder.

* Free convection cooling may become important (for the removal of fission-product decay heat) in other reactors after shut-down when the forced flow of coolant is stopped.

11.150. Since equation (11.149.1) is applicable only for a single heat-transfer surface, it is not suitable for application to reactors of the Swimming Pool type. A method for treating such systems is described in § 11.165, and other discussions will be found in the literature.*

PRESSURE DROPS AND PUMPING POWER IN COOLANT SYSTEMS

Pressure Drop Due to Friction

11.151. Pumping power requirements of a reactor are determined by the pressure drop in the cooling system and the rate of flow of the coolant. The total pressure drop for an incompressible fluid is the sum of losses due to friction in channels, pipes, and fittings, and to changes in the cross-sectional flow area. Although these topics are more adequately treated in the standard engineering texts,† they will be reviewed here briefly.

11.152. The pressure drop, Δp_f, accompanying isothermal *laminar* flow at a mean velocity v in a circular pipe of diameter D and length L is given by the familiar Poiseuille equation

$$\Delta p_f = \frac{32\mu v L}{gD^2},$$

which may be written

$$\Delta p_f = 64\,\frac{\mu}{Dv\rho}\cdot\frac{\rho v^2 L}{2gD}, \qquad (11.152.1)$$

where μ is the viscosity and g the acceleration of gravity.‡ The subscript f is used in Δp_f to indicate that the pressure loss described is due to fluid friction. It will be observed that the factor $\mu/Dv\rho$ on the right of equation (11.152.1) is the reciprocal of the Reynolds number (§ 11.119), and so

$$\Delta p_f = \frac{64}{\mathrm{Re}}\cdot\frac{\rho v^2 L}{2gD}. \qquad (11.152.2)$$

This gives the pressure drop due to fluid friction for laminar flow in a straight pipe of circular cross section.

11.153. For turbulent flow at a mean velocity v, the relationship corresponding to equation (11.152.2) is

$$\Delta p_f = 4f\,\frac{\rho v^2 L}{2gD}, \qquad (11.153.1)$$

* D. C. Hamilton, *et al.*, Reports ORNL-1624, ORNL-1769; D. V. Wordsworth, Report AERE E/R 1270.

† See, for example, McAdams, *op. cit.*; G. G. Brown (Ed.), "Unit Operations," John Wiley & Sons, 1950.

‡ This is, strictly, the gravitational constant for relating mass and force, but the distinction between this constant and the acceleration of gravity will be ignored here.

known as the Fanning equation, in which the dimensionless quantity f is called the *friction factor*.* Comparison of equations (11.152.2) and (11.153.1) shows that for laminar flow $f = 16/\mathrm{Re}$, but the relationship between the friction factor and the Reynolds number in the case of turbulent flow is more complicated. Several empirical expressions have been developed; one of these, which holds with a fair degree of accuracy for flow in smooth pipes at Reynolds numbers up to about 2×10^5, is the Blasius equation†

$$f = 0.079\ \mathrm{Re}^{-0.25}. \qquad (11.153.2)$$

This permits the evaluation of the turbulent-flow friction factor for a given Reynolds number.

11.154. For turbulent flow in a commercial rough pipe the friction factor is larger than for a smooth pipe at the same Reynolds number, as shown in Fig. 11.18. The deviation from the ideal behavior of a smooth pipe increases with

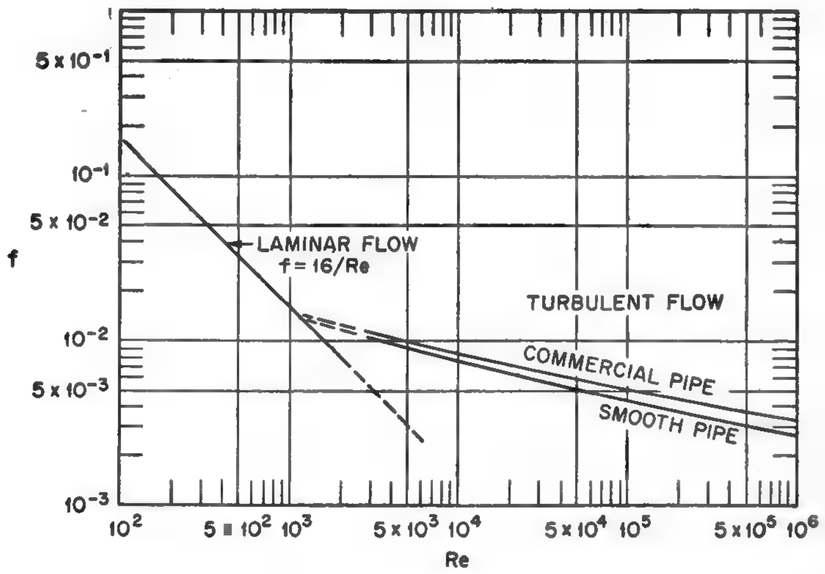


FIG. 11.18 Friction factor for smooth and commercial pipes

the roughness of the pipe, especially at high Re values. The variation of the friction factor with Reynolds number has been determined experimentally for different degrees of roughness, expressed in terms of a dimensionless quantity ϵ/D, where ϵ is a measure of the size of the roughness projections and D is the pipe diameter.‡

11.155. For turbulent flow in noncircular channels of relatively simple form, the pressure drop due to friction may be calculated by substituting the equiv-

* In the equivalent Darcy-Weisbach equation a friction factor equal to $4f$ is used.
† H. Blasius, 1913.
‡ Cf., L. F. Moody, *Trans. Am. Soc. Mech. Engrs.*, **66**, 671 (1944).

alent diameter, defined in § 11.122, for D in equation (11.153.1). The same value is used in determining the Reynolds number. It may be mentioned that the replacement of D by D_e for noncircular channels is incorrect for laminar flow.

Example Estimate the pressure in lb/in.² (psi) required to overcome friction for the turbulent flow of water in the channel between the fuel plates in the examples given earlier.

The value of Re was found in § 11.122 to be 8050 and so f, assuming the plates to be moderately smooth, is seen from Fig. 11.18 to be about 0.008. Since ρ is 61.8 lb/ft³, v is 10,800 ft/hr, L is 2.5 ft, D_e is 0.02 ft and g is 32.2 ft/sec² = (32.2)(3600)² ft/hr², it follows from equation (11.153.1) that

$$\Delta p_f = (4)(0.008)\frac{(61.8)(10{,}800)^2(2.5)}{2[(32.2)(3600)^2](0.0833)}$$
$$= 34.5\ \mathrm{lb/ft^2}$$
$$= 0.24\ \mathrm{lb/in.^2\ (psi)}.$$

Pressure Drop Due to Changes in Velocity and Direction

11.156. In a flowing fluid there will be changes in pressure due to changes in velocity resulting from gradual or abrupt changes in flow area. Such pressure changes are usually considered in terms of the "velocity head," defined as $v^2/2g$; the pressure corresponding to a head H is equal essentially to $H\rho$. Since v^2 is generally high for turbulent flow, the pressure losses accompanying changes in cross-sectional area of fluid conduits may be very significant.

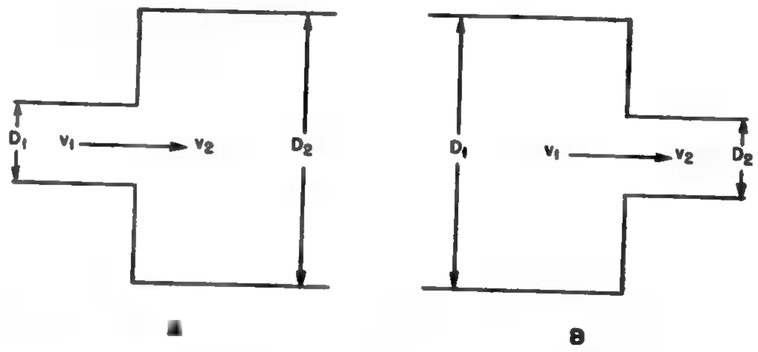


FIG. 11.19 A. Abrupt expansion in fluid flow
B. Abrupt contraction in fluid flow

11.157. In general, for abrupt expansion (Fig. 11.19, A) or contraction (Fig. 11.19, B) the loss of head, ΔH, which occurs in either case, can be represented by

$$\Delta H\ (\text{expansion}) = K_e\frac{v_1^2}{2g}$$

$$\Delta H\ (\text{contraction}) = K_c\frac{v_2^2}{2g},$$

where v_1 and v_2 are the upstream and downstream velocities, respectively; the value of K, the loss coefficient, depends upon the conditions, as indicated below.*

11.158. For abrupt *expansion*,

$$K_e = \left(1 - \frac{D_1^2}{D_2^2}\right)^2,$$

where D_1 and D_2 are the pipe diameter upstream and downstream, respectively. For expansion into a large reservoir, D_2 is large, and K_e becomes virtually unity. The loss of head is then almost equal to $v_1^2/2g$.

11.159. For abrupt *contraction* the value of K_c varies with the ratio D_2/D_1 in the following manner:

D_2/D_1	0.8	0.6	0.4	0.2	0
K_c	0.13	0.28	0.38	0.45	0.50

For the case of inlet from a very large reservoir, D_2/D_1 approaches zero, and K_c is then approximately 0.5; the corresponding loss of head is $0.5v_2^2/2g$.

11.160. It should be understood that the results given above apply only to cases of abrupt expansion or contraction. The loss of head decreases if the fluid exit is rounded, making the change less abrupt. Where the exit is tapered so that the included angle is 7° or less, the losses will usually be negligible.

Example Estimate the pressure loss due to contraction and expansion of coolant entering or leaving a channel between two parallel-plate fuel elements considered previously.

The coolant undergoes sudden contraction and expansion as it enters and leaves the channel, respectively, from or to a header or manifold of large cross section. The loss of head upon entering the channel, i.e., upon contraction, may then be taken as $0.5v_2^2/2g$, where v_2 is here the velocity of the coolant in the channel. Similarly, upon leaving the channel, the loss is $v_1^2/2g$, where v_1 is also the velocity of the coolant in the channel. The total loss of head upon entry and exit is thus $1.5v^2/2g$, where v is the velocity of the coolant in the channel, i.e., 10,800 ft/hr. The pressure loss is equal to the loss of head multiplied by the density of the coolant (§ 11.156); hence,

$$\begin{aligned}\text{Pressure loss} &= \frac{(1.5)(10{,}800)^2(61.8)}{2[(32)(3600)^2]} \\ &= 13.0 \text{ lb/ft}^2 \\ &= 0.090 \text{ psi.}\end{aligned}$$

11.161. Losses in pipe fittings, due to changes in direction, e.g., in elbows, curves, etc., or to contraction in valves, can also be expressed in terms of the velocity head, e.g., $K_f v^2/2g$. The factor K_f may range from 0.25 or less for a gradual 90° curve or a fully opened gate valve to 1.0 for a standard screwed 90° elbow, or as high as 10 for a fully opened globe valve.†

* Cf. V. L. Streeter, "Fluid Mechanics," McGraw-Hill Book Co., Inc., 1951, p. 213; H. Rouse and J. W. Howe, "Basic Mechanics of Fluids," John Wiley and Sons, Inc., 1953, p. 153.

† For further information, standard reference books and manufacturers' catalogs should be consulted. The loss coefficients, K_f, are often expressed in terms of an "equivalent" length of straight pipe.

Pumping Power

11.162. The work required to pump coolant through a reactor is given by

$$\begin{aligned}\text{Pumping power} &= \text{Pressure drop} \times \text{Volume flow rate} \\ &= (\Delta p)(A_f v),\end{aligned} \qquad (11.162.1)$$

where Δp is the total pressure drop due to all causes and A_f is the total cross-sectional (or flow) area of all the coolant channels. Since flow is ordinarily turbulent, where forced convection heat transfer is employed, the pressure drop due to friction is given by equation (11.153.1); and to this must be added the values for entrance and exit effects, given above, as well as for various fittings, etc., in the coolant circuit.

11.163. Attempts have been made to compare the pumping power, relative to the total reactor power, required to overcome the pressure drop due to friction for various coolants.* This has been done by combining equations (11.67.1), (11.125.1) and (11.153.1), together with the definition of Re, and the relationship of equation (11.137.1). In making such comparisons, however, it is necessary to postulate that various conditions in the reactor are independent of the nature of the coolant. Thus, it is supposed that, for a given reactor power, the temperature rise, Δt_c, of the coolant is always the same. But this may not necessarily be true in practice for optimum operation. For example, using a gaseous coolant, it may be desirable to have Δt_c much larger than for a liquid metal. Further, the comparisons of pumping power are based on overcoming fluid friction in the reactor only and do not take into account the heat exchanger, as well as entrance and exit effects which may be considerable.

11.164. Without being quantitative, however, it is possible to draw some general conclusions with regard to pumping power. It is seen that the pressure losses due to fluid friction as well as to sudden changes in flow area all increase with v^2. Further, it was stated in § 11.137 that, because the volumetric heat capacity of a gaseous coolant is normally much less than of a liquid, the removal of a certain quantity of heat will usually require a very much higher coolant velocity for a gas than for a liquid, assuming that $A_f \Delta t_c$ is not greatly different in the two cases. Hence the pumping power requirements for a gas will be much larger than for a liquid coolant. The discrepancy can be partially overcome by increasing the density of the gas, i.e., by operating at high pressures (cf. § 8.232).

Example Estimate the power required for pumping the cooling water through the core of the 3000-kw reactor considered in previous examples, assuming the minimum number of cooling channels to be 450.

The pressure drop due to fluid friction and entrance and exit effects has been found to be 34.5 + 13 = 47.5 lb/ft². The total flow area of the 450 channels is

* For example, E. R. Gilliland, *et al.*, see reference to § 11.61; Report on Atomic Power Symposium, Chalk River, Ontario, 1953 (Report CRR-548-A), Chapter X (by J. B. Haddow). See also, P. W. Parsons and B. J. Gaffney, *Trans. Am. Inst. Chem. Engrs.*, **40**, 655 (1944); J. J. Carberry, *Chem. Eng.*, **61**, 225 (1953).

$$A_f = \frac{(450)(2.5)(0.12)}{144} = 0.94 \text{ ft}^2.$$

The velocity of the coolant is 10,800 ft/hr so that

$$\text{Pumping power} = (47.5)(0.94)(10{,}800)$$
$$= 4.82 \times 10^5 \text{(ft)(lb)/hr.}$$

Converting to theoretical horsepower, making no allowance for pump efficiency,

$$\text{Pumping power} = \frac{4.82 \times 10^5}{(33{,}000)(60)} = 0.243 \text{ hp.}$$

(The total pressure drop for the entire cooling system, including piping and heat exchangers, will be considerably larger than the value used here and thus the total pumping power will also be greater.)

Pressure Drop and Free Convection Cooling

11.165. Valuable information concerning the cooling of reactors of the Swimming Pool type by free convection can be obtained from pressure drop considerations. The flow of water between the parallel-plate elements in these reactors is approximately laminar, because of the close spacing of the plates, the low heat flux, and the consequent small convective driving force. The flow rate and, hence, the ability to carry heat from the core may be estimated in the following manner.

11.166. For laminar flow between parallel plates, the appropriate form of the Poiseuille equation leads to the expression

$$\Delta p_f = \frac{48\mu v L}{g D_e^2} \tag{11.166.1}$$

for the pressure drop through the entire length L of the fuel plate; as before, D_e is the equivalent diameter. For convective cooling of a vertical plate, v in equation (11.166.1) will then be the average upward velocity of the coolant when Δp_f is equal to the average pressure drop due to the difference in density at bottom and top. If ρ_1 and ρ_2 are the respective densities of the coolant at entrance and discharge, respectively, from the channel, the average pressure differential is approximately $\frac{1}{2}(\rho_1 - \rho_2)L$. If this is equated to the right side of equation (11.166.1), the average velocity of the fluid is given by

$$v_{\text{av}} = \frac{(\rho_1 - \rho_2) g D_e^2}{96\mu_{\text{av}}}, \tag{11.166.2}$$

where μ_{av} is the average viscosity.

11.167. The total rate of heat production by the reactor, q_{tot}, is obtained from the general equation (11.137.1). In the present case, v is given by equation (11.166.2), ρ is the mean density, i.e., $\frac{1}{2}(\rho_1 + \rho_2)$, and Δt_c is $t_2 - t_1$, the temperature difference between top and bottom. Hence,

$$q_{\text{tot}} = (\rho_1 + \rho_2)(\rho_1 - \rho_2) \frac{A_f g D_e^2 c_p (t_2 - t_1)}{192\mu_{\text{av}}},$$

which may be used to estimate the power output for specified lower and upper temperatures.

Example In a free-convection cooled Swimming Pool reactor, the plates are 0.01 ft apart and the total flow area for the water is 0.75 ft². The temperature of the water at the bottom of the fuel plates is 80°F. Estimate the power output when the upper temperature is (*a*) 100°F and (*b*) 120°F, using the following data:

t °F	ρ lb/ft³	μ lb/(ft)(hr)	c_p Btu/(lb)(°F)
80°	62.21	2.08	1.00
100°	62.00	1.66	1.00
120°	61.73	1.36	1.00

(*a*) Upper temperature = 100°F. Taking the average viscosity as the arithmetic mean for the lower and upper temperatures,

$$q_{\text{tot}} = \frac{(124.21)(0.21)(0.75)[(32)(3600)^2](0.02)^2(1.00)(20)}{(192)[(\frac{1}{2})(2.08 + 1.66)]} \text{ Btu/hr}$$
$$= 53 \text{ kw.}$$

(*b*) Upper temperature = 120°F

$$q_{\text{tot}} = \frac{(123.94)(0.48)(0.75)[(32)(3600)^2](0.02)^2(1.00)(40)}{(192)[(\frac{1}{2})(2.08 + 1.36)]} \text{ Btu/hr}$$
$$= 260 \text{ kw.}$$

11.168. In reactors of the Swimming Pool type, with free convection cooling, higher power levels than those calculated here are possible by operating at higher exit-water temperatures. However, at power levels above about 200 kw other factors, such as radioactivity of the pool water and surface temperature of the fuel elements, must be taken into consideration. These may limit the total power of reactors of this type.

11.169. It should be pointed out that, in the foregoing analysis, no consideration has been given to the existence of the heated jet of liquid rising in a column above the reactor core. That an additional driving force due to this column of liquid does exist has been proved by experiment, and it may be more important in promoting flow of coolant through the reactor than the vertical driving force taken over the length of the fuel plates. Other effects which may be significant, including pressure losses at channel inlet and discharge, deviations from laminar flow, and nonuniform surface temperatures, have also been neglected.

HEAT TRANSFER TO BOILING LIQUIDS*

Surface and Volume Boiling

11.170. Boiling is of importance in nuclear reactor systems both as a means of achieving high heat-transfer rates and of generating steam. The use of a boiling coolant within the core is applicable to reactors employing either solid or liquid fuels. In a solid-fuel reactor, heat is transferred by "surface boiling"; heat generated in the solid fuel must flow to the coolant across a solid-fluid interface at which bubble development occurs. Similar considerations apply to the generation of steam in a secondary coolant system. In a reactor with a boiling liquid fuel, "volume boiling" occurs; fuel distributed throughout the coolant, usually as a solution, will cause bubble formation within the bulk of the coolant itself.

11.171. For the treatment of *surface boiling* it is convenient to consider an element of heated surface at temperature t_s, in contact with a liquid at an average temperature t_f. Suppose the temperature difference between the heated surface and the liquid bulk, i.e., $t_s - t_f$, is steadily increased; the corresponding variation in the heat flux, i.e., q/A, across the surface is then as shown in Fig. 11.20.

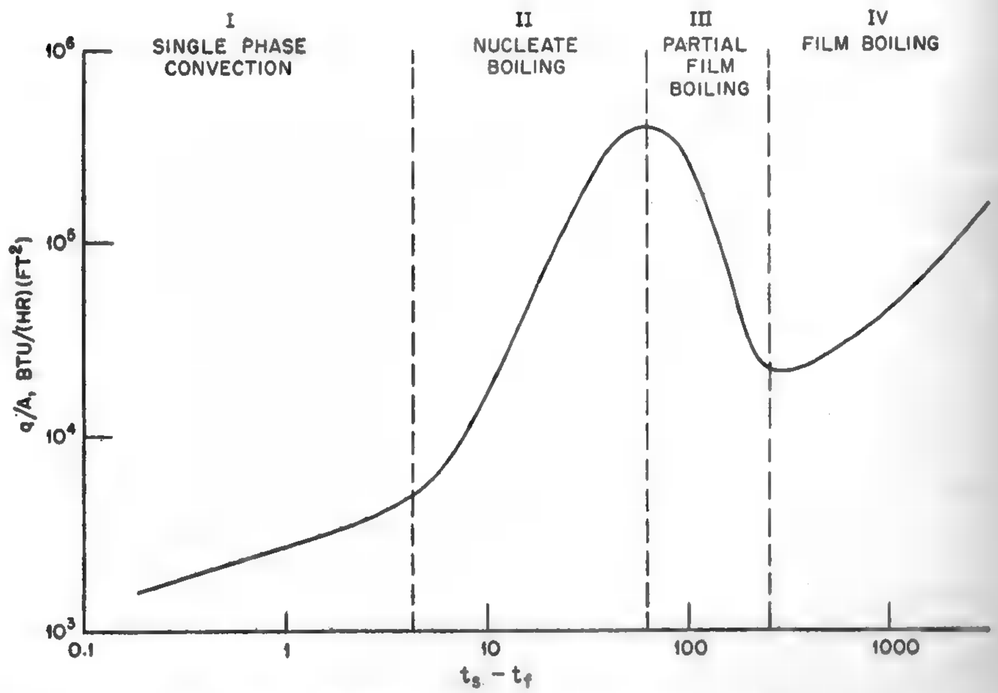


FIG. 11.20 Variation of heat flux with surface-liquid temperature difference in boiling system

* For general discussion and references, see W. H. McAdams, *op. cit.* (3rd ed.), Chapter 14; M. Jakob, *op. cit.*, Chapter 29.

Although the data in this figure are representative of natural convection boiling from a heated wire in a pool of water at atmospheric pressure and a liquid temperature of 212°F, some of the same general characteristics apply to forced convection boiling and other pressure and temperature conditions.

11.172. The curve can be divided into a number of regions, in each of which the mechanism of heat transfer is somewhat different from that in the others. Until the heated surface exceeds the saturation temperature* by a small amount, heat is transferred by single-phase convection; this occurs in region I. The system is heated by slightly superheated liquid rising to the liquid-pool surface where evaporation occurs. In region II, vapor bubbles form at the heated surface; this is the *nucleate boiling* range in which formation of bubbles occurs upon nuclei, such as solid particles or gas adsorbed on the surface, or gas dissolved in the liquid. Nucleate boiling is a common phenomenon, since it is encountered in standard power-plant steam generators.

11.173. The steep slope of the heat flux curve in region II is due to the mixing of the liquid resulting from the action of the vapor bubbles. A maximum flux is attained when the bubbles become so dense that they coalesce and form a vapor film over the heating surface. The heat must then pass through the vapor film by a combined mechanism of conduction and radiation, neither of which is particularly effective in this temperature range. Consequently, beyond the maximum, the heat flux decreases appreciably in spite of an increase of temperature. In region III, the film is unstable; it spreads over a part of the heating surface and then breaks down. Under these conditions violent nucleate boiling occurs at some portions of the surface, while *film boiling*, due to heat transfer by conduction and radiation, occurs at other portions.

11.174. For sufficiently high values of $t_s - t_f$, as in region IV, the film becomes stable, and the entire heating surface is covered by a thin layer of vapor; boiling is then exclusively of the film type. If attempts are made to attain large heat fluxes with film boiling, as high as those possible with nucleate boiling, for example, the temperature of the heated surface may become so high as to result in destruction of the heater. This is called a "burn-out" and is, of course, to be avoided. It may be noted, too, from Fig. 11.20, that if a system undergoing nucleate boiling is operating at conditions near the maximum of the curve, a slight increase in the heat flux will cause a sudden change to film boiling, which usually results in burn-out.

11.175. *Subcooled boiling*† occurs when the bulk temperature of the liquid is below saturation but that of the surface is above saturation. Vapor bubbles form at the heating surface but condense in the cold liquid, so that no net generation of vapor is realized. Very high heat fluxes can be obtained under these conditions; values as large as 1.4×10^7 Btu/(hr)(ft²) have been reported in

* The saturation temperature is the temperature of the saturated vapor, i.e., saturated steam, at the existing pressure.

† Sometimes called local or surface boiling in the general literature.

forced-convection heating of water, but 2×10^6 Btu/(hr)(ft^2) appears more realistic if the surface temperatures are to be kept low enough to avoid burn-out.

11.176. In *volume boiling* the heat is generated within the liquid and only ordinary nucleate boiling occurs. Hence, with volume boiling, burn-out problems are eliminated. The boiling liquid temperature is very slightly higher than the saturation value and is essentially independent of the rate of heat generation, i.e., of the power density.

Boiling in Reactor Systems

11.177. Suppose that water, below the saturation temperature, is forced through a channel between or around the solid fuel elements of a reactor; heat is then transferred from the solid surface to the water. As long as the fuel-wall temperature, which increases along its length in the manner described in § 11.102, *et seq.*, remains below the steam saturation temperature, single-phase heat transfer only will occur. This is the condition aimed at in most reactors employing water as coolant or as combined moderator and coolant. In some instances, such as the Submarine Thermal Reactor (STR) and the Pressurized Water Reactor (PWR), the pressure of the coolant system is increased in order to raise the saturation temperature and thus prevent boiling.

11.178. In boiling reactors (§§ 8.238, 12.149), however, the wall temperature, t_s, is permitted to exceed the saturation temperature, t_{sat}. At first there will be

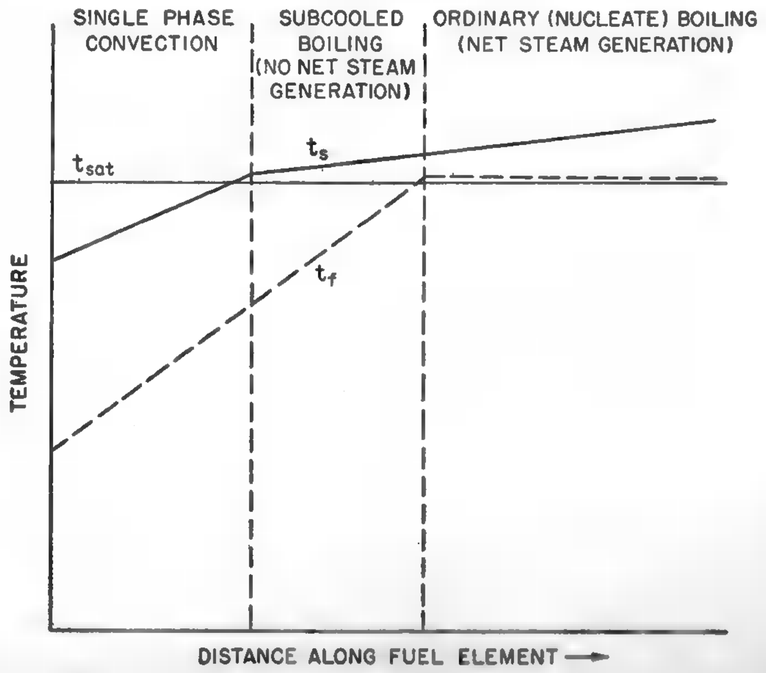


Fig. 11.21 Variation of surface and liquid temperatures with distance from entry in boiling system

subcooled boiling, while the bulk coolant temperature, t_f, is below saturation, and this will be followed by ordinary nucleate boiling with net steam generation (Fig. 11.21). In this region the rate of heat transfer is high, and the conditions, e.g., coolant flow rate and inlet temperature, should be chosen so as to maintain nucleate boiling over almost the whole length of the fuel element.

11.179. If, due to local circumstances, the temperature difference between a point (or region) on the fuel-element surface and the water is increased, film boiling may take place. There is then a danger that burn-out will occur. This is most likely to happen near the exit of the coolant channel where the temperature of the metal surface is a maximum (§ 11.112). If two or more channels are connected in parallel, as is usually the case, the danger of burn-out is greatly increased, for the following reason. The formation of a film or layer of steam on the metal in a particular channel will result in an increased pressure drop over the length of that channel. When several channels are fed from a common header, the flow of water in the channel under consideration will then decrease, since it offers the largest resistance. This will tend to increase steam-film formation, which will be accompanied by a further decrease in flow, and so on until burn-out occurs.

11.180. In addition to the production of steam from water in a reactor as described above, or by passage of water through tubes immersed in a homogeneous fuel solution, which is somewhat similar, there is a possibility of steam formation by direct boiling of such an aqueous solution. In systems of this type the heat generation rate is strongly dependent upon the density of the boiling water-steam mixture, since the liquid coolant usually functions also as moderator, as well as the fuel carrier. The most serious consequence of a decrease in density is usually an increase in the neutron leakage.

11.181. Vapor generated in the boiling liquid in the interior of the reactor will rise as a result of its buoyancy. In addition to this natural rise of the vapor, the gross motion or circulation of the liquid can contribute to the transport of vapor. A natural circulation can be achieved if the flow can be directed to take advantage of the variation of fluid density within the core. If necessary, forced circulation, in which mechanical energy is supplied to maintain the desired fluid motion by an agency external to the core, e.g., a pump, may be employed.

11.182. The calculation of the mixture density in two-phase flow for both heterogeneous and homogeneous systems is complicated by the fact that the liquid and vapor normally possess different average velocities because of the slip of the vapor relative to the liquid. However, it is possible to make a gross evaluation of the flow conditions.*

Boiling Heat-transfer Coefficients

11.183. It is apparent from Fig. 11.20 that log (q/A) is a linear function of log $(t_s - t_f)$ over a considerable portion of the nucleate boiling range, so that the

* Cf., R. C. Martinelli and D. B. Nelson, *Trans. Am. Soc. Mech. Engrs.*, 70, 695 (1948); see also, S. Untermyer, *Nucleonics*, 12, No. 7, 48 (1954).

general expression

$$\frac{q}{A} = c(t_s - t_f)^n, \tag{11.183.1}$$

where c and n are constants, is applicable to a particular set of conditions. Since t_f is essentially the same as the saturation temperature (see Fig. 11.21), $t_s - t_f$ is virtually equal to the difference between the temperature of the heated surface and the saturation temperature of the coolant; this difference may be represented by Δt_b. Hence, equation (11.183.1) may be written as

$$\frac{q}{A} = c(\Delta t_b)^n.$$

If h_b represents the boiling heat-transfer coefficient, it may be defined by

$$\frac{q}{A} = h_b \Delta t_b, \tag{11.183.2}$$

so that

$$h_b = c(\Delta t_b)^{n-1}.$$

11.184. Curves showing h_b in Btu/(hr)(ft²)(°F) as a function of Δt_b in °F, obtained from a variety of experimental studies of heat transfer to boiling water in a free convection system at several pressures, are given in Fig. 11.22.* The value of the exponent $n - 1$ is found to be 1.42 in all cases, but the factor c increases with the pressure; thus,

$$h_b = c(\Delta t_b)^{1.42}, \tag{11.184.1}$$

with the following values of c:

Pressure, psia	14.7 (1 atm)	215	370	520
c	42	82	90	100

These results are applicable over a range of Δt_b from about 1° to 45°F.

Example Determine the boiling heat-transfer coefficient of water at a pressure of 215 psia, when the temperature of the metal at the interface is 25°F above saturation.

In this case, Δt_b is 25°F and c is 82; hence, from equation (11.184.1),

$$h_b = (82)(25)^{1.42}$$
$$= 7500 \text{ Btu/(hr)(ft}^2\text{)(°F)}.$$

(High heat-transfer rates are evidently possible from a surface at which nucleate boiling of water occurs even for moderate temperatures above saturation.)

Heat Exchange to Steam System

11.185. The design of the heat exchanger for transferring heat from the primary to a secondary coolant without boiling follows conventional engineering practice as far as the heat-transfer calculations are concerned. Because of induced radioactivity in most coolants, however, more rigid specifications with regard to construction, in order to avoid leakage and minimize corrosion and other factors causing maintenance problems, are imposed than is usual for commercial equipment. In addition, the need for locating the heat exchanger behind a shield may affect the design.

* Based upon data in the published literature; see, for example, M. T. Cichelli and C. F. Bonilla, *Trans. Am. Inst. Chem. Engrs.*, **41**, 755 (1945).

11.186. The operation of nuclear power plants, with the exception of those using boiling reactors, involves the production of steam by transfer of heat from the primary or secondary coolant to water. The design of the heat exchanger or steam generator is thus an essential aspect of the over-all reactor heat-transfer system problem. This involves a somewhat unique application of boiling heat-transfer data.*

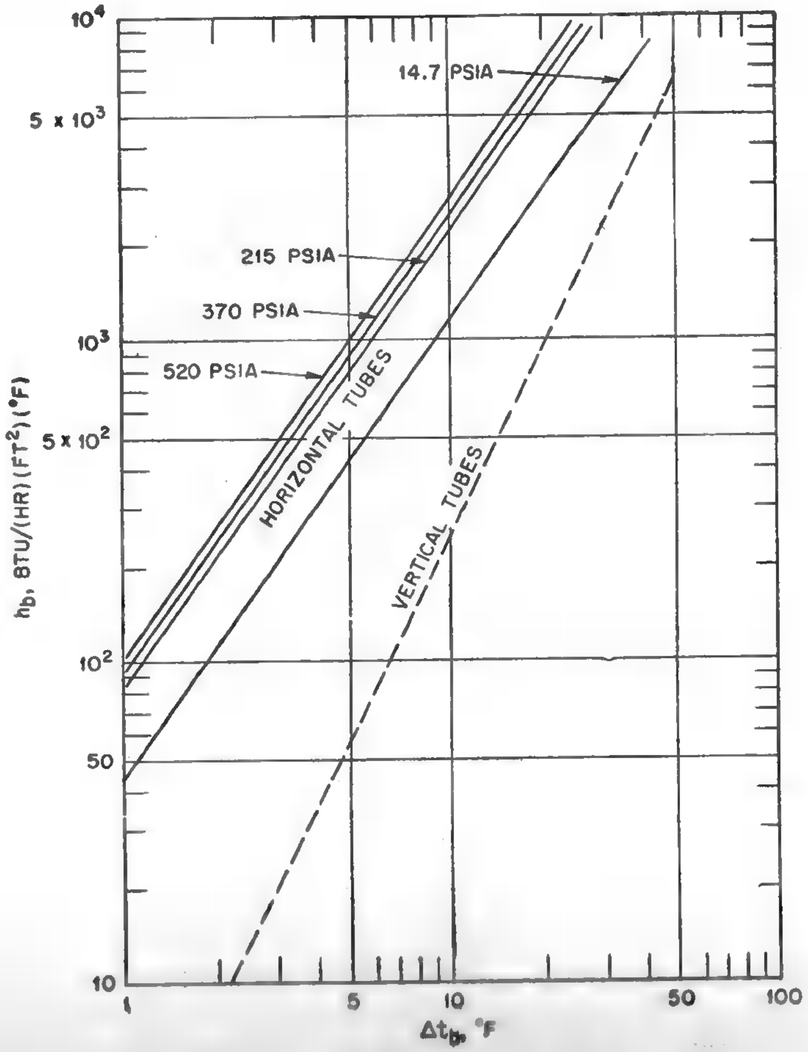


Fig. 11.22 Boiling heat-transfer coefficient

* J. A. Lane, unpublished.

11.187. Consider a steam generator in which the reactor coolant, of specific heat c_p, flows through a tubular heat exchanger, at a mass flow rate w, and transfers heat to boiling water outside the tubes. The rate of heat loss from the coolant in a differential section of the steam generator is then given by

$$dq = wc_p\, d(\Delta t), \tag{11.187.1}$$

where Δt is the total temperature difference between the reactor coolant and saturated steam in the region of the heat exchanger being considered. The same heat-loss rate can be expressed in terms of the over-all heat-transfer coefficient, U, of the heat exchanger (cf. § 11.71); thus, the heat transferred from the coolant to the boiling water is

$$dq = U\Delta t\, dA, \tag{11.187.2}$$

where dA is the heat-transfer area of the differential section. Upon equating the two expressions for dq, and integrating between the limits of Δt_1 and Δt_2, where the subscripts 1 and 2 refer to the two ends of the steam-generating heat exchanger, it is found that

$$A = wc_p \int_{\Delta t_1}^{\Delta t_2} \frac{d(\Delta t)}{U\Delta t}, \tag{11.187.3}$$

where A is the total heat-transfer area; w and c_p are assumed to be independent of Δt.

11.188. The temperature difference Δt can be regarded as being made up of three parts (Fig. 11.23), viz.,

$$\Delta t = \Delta t_s + \Delta t_w + \Delta t_b,$$

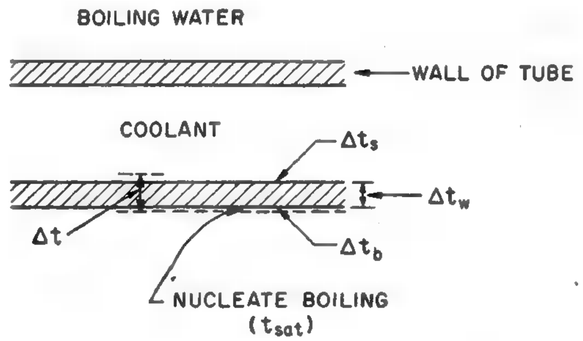


FIG. 11.23 Temperature differences in a steam generator

where Δt_s is the difference between the mixed-mean fluid (coolant) temperature and the interior wall surface of the heat-exchanger tube, Δt_w is the temperature drop across the wall of the tube, and Δt_b is the temperature difference between the outer wall and the steam saturation temperature, as defined in § 11.183. These three temperature differences may be expressed in terms of the heat-flow rate, the area A, and the appropriate heat-transfer coefficients or thermal conductances.

11.189. If a is the inner radius and b the outer radius of the heat-exchanger tube, then it follows from the equations in § 11.73, *et seq.*, that

$$\Delta t_s = \frac{q}{A} \cdot \frac{1}{h}\left(\frac{b}{a}\right),$$

where A is the heat-transfer area based on the *outside diameter* of the tube and h is the heat-transfer coefficient of the coolant inside the tube. Similarly, it can be readily shown that

$$\Delta t_w = \frac{q}{A}\left[\frac{b \ln (b/a)}{k}\right],$$

where k is the thermal conductivity of the tube wall, and

$$\Delta t_b = \frac{q}{A} \cdot \frac{1}{h_b},$$

where h_b is the apparent boiling heat-transfer coefficient (§ 11.183). Upon substituting the expression for h_b given by equation (11.184.1) and summing the three terms for the separate temperature differences, it is found that

$$\Delta t = \left[\frac{b \ln (b/a)}{k} + \frac{b}{ah}\right]\frac{q}{A} + \frac{1}{c^{0.413}}\left(\frac{q}{A}\right)^{0.413}. \tag{11.189.1}$$

11.190. This equation gives the relation between the over-all temperature drop, Δt, i.e., from the coolant inside the tubes to the saturated steam outside of tubes, and the heat flux, q/A, based on the outside area. If it were possible to solve this equation explicitly for q/A, the resulting expression could be substituted for $U\Delta t$ in equation (11.187.3) and the integration performed. Since this is not possible, an alternative approach is used. From equations (11.187.1), (11.187.3), and (11.189.1) it is found that

$$\begin{aligned} A &= wc_p \int_{(q/A)_2}^{(q/A)_1} \frac{\left[\frac{b \ln (b/a)}{k} + \frac{b}{ah}\right] d\left(\frac{q}{A}\right) + \frac{0.413}{c^{0.413}}\left(\frac{q}{A}\right)^{-0.587} d\left(\frac{q}{A}\right)}{q/A} \\ &= wc_p\left[\frac{b \ln (b/a)}{k} + \frac{b}{ah}\right]\int_{(q/A)_2}^{(q/A)_1} d \ln \frac{q}{A} + wc_p \frac{0.413}{c^{0.413}} \int_{(q/A)_2}^{(q/A)_1} \frac{d(q/A)}{(q/A)^{1.587}}. \\ &= wc_p\left[\frac{b \ln (b/a)}{k} + \frac{b}{ah}\right] \ln \frac{(q/A)_1}{(q/A)_2} + \frac{0.705wc_p}{c^{0.413}}\left[\left(\frac{q}{A}\right)_2^{-0.587} - \left(\frac{q}{A}\right)_1^{-0.587}\right], \end{aligned} \tag{11.190.1}$$

where the subscripts 1 and 2 refer to the conditions at the coolant entrance and exit, respectively, of the heat exchanger (or steam generator).*

* For coolant velocities of 5 to 25 ft/sec in small tubes and steam pressures of 100 to 500 psia, a check on the calculations may be obtained from the approximate relationship

$$A = \frac{wc_p}{80}\left[\frac{1}{(\Delta t_2)^{0.3}} - \frac{1}{(\Delta t_1)^{0.3}}\right].$$

Example A pressurized-water reactor operates at a (heat) power level of 30,000 kw. Cooling water enters the reactor at 482°F and leaves at 572°F and 2000 psia. In the stainless-steel tubular heat exchanger (boiler), through which the cooling water flows at 10 ft/sec, saturated steam is produced at 471°F and 520 psia, outside the tubes. The latter are 0.375 in. O.D. and 0.245 in. I.D. Calculate (*a*) the heat-transfer area of the heat exchanger in which steam is generated and (*b*) the number and length of the heat-exchanger tubes. The following data may be used for water at 527°F, the average temperature in the reactor:

$$c_p = 1.024 \text{ Btu/(lb)(°F)} \qquad \rho = 50 \text{ lb/ft}^3.$$

For Type 347 stainless steel

$$k = 10.8 \text{ Btu/(hr)(ft}^2\text{)(°F/ft)}.$$

Since h for water does not vary greatly in the temperature range of interest, it may be calculated at the average temperature, 527°F. From equation (11.128.1), with $v = 10$ ft/sec and $D = 0.245$ in., it follows that

$$h = 170[1 + (10^{-2})(527) - (10^{-5})(527)^2] \frac{(10)^{0.8}}{(0.245)^{0.2}}$$
$$= 5000 \text{ Btu/(hr)(ft}^2\text{)(°F)}.$$

The internal radius, a, of the heat-exchanger tubes is $(\frac{1}{2})(0.245) = 0.125$ in. $= 0.0102$ ft, and the external radius, b, is $(\frac{1}{2})(0.375) = 0.1875$ in. $= 0.0156$ ft: hence,

$$\frac{b \ln (b/a)}{k} = \frac{0.0156}{10.8} \ln \frac{0.0156}{0.0102}$$
$$= 0.00061.$$

Further,

$$\frac{b}{ah} = \frac{0.0156}{(0.0102)(5000)} = 0.00031.$$

The heat-generation rate in the reactor is 30,000 kw or $(30{,}000)(3412) = 1.024 \times 10^8$ Btu/hr; this is equal to q in the equation (cf. § 11.108)

$$q = wc_p\Delta t_c,$$

where Δt_c is the increase in the coolant temperature in the reactor, i.e., $572 - 482 = 90$°F. Hence,

$$wc_p = \frac{q}{\Delta t_c} = \frac{1.024 \times 10^8}{90}$$
$$= 1.14 \times 10^6 \text{ Btu/(hr)(°F)}$$

and, since c_p is 1.024 Btu/(lb)(°F),

$$w = \frac{1.14 \times 10^6}{1.024} = 1.11 \times 10^6 \text{ lb/hr}.$$

In order to evaluate $(q/A)_1$ and $(q/A)_2$ at entrance and exit, respectively, of the heat exchanger, use is made of equation (11.189.1). The values of the expressions containing a, b, h, and k have already been determined, and from the data in § 11.184 it is seen that at a steam pressure of 520 psia the constant c is equal to 100. Consequently equation (11.189.1) becomes

$$\Delta t = (0.00061 + 0.00031)\frac{q}{A} + \frac{1}{(100)^{0.413}}\left(\frac{q}{A}\right)^{0.413}.$$

From this relationship it is possible to make a plot of q/A against Δt for various values of q/A. From this plot it is found that at the coolant entrance to the heat exchanger, where Δt_1 is $572 - 471 = 101$°F,

$$(q/A)_1 = 92{,}000 \text{ Btu/(hr)(ft}^2\text{)},$$

and at the exit, where Δt_2 is $482 - 471 = 11$°F,

$$(q/A)_2 = 6050 \text{ Btu/(hr)(ft}^2\text{)}.$$

(The assumption is made here that the saturation temperature of the water is the same at both ends of the heat exchanger.)

(*a*) The total (outside) area of the heat-transfer tubes can now be obtained from equation (11.190.1); thus,

$$A = (1.14 \times 10^6)(0.00061 + 0.00031) \ln \frac{92{,}000}{6050}$$
$$+ \frac{(0.705)(1.14 \times 10^6)}{(100)^{0.413}} [(6050)^{-0.587} - (92{,}000)^{-0.587}].$$
$$= 3380 \text{ ft}^2.$$

By means of the approximate equation in the footnote to § 11.190, it is found that

$$A = \frac{1.14 \times 10^6}{80}\left[\frac{1}{(11)^{0.2}} - \frac{1}{(101)^{0.2}}\right]$$
$$= 3170 \text{ ft}^2,$$

which is in satisfactory agreement with the more exact value.

(*b*) The number of tubes, n, in the heat exchanger is obtained from the relationship

$$w = nv\rho A_f,$$

where A_f is the flow area per tube. In the present case w is 1.11×10^6 lb/hr, v is 10 ft/sec or 3.6×10^3 ft/hr, ρ is 50 lb/ft^3, and the flow area πa^2 is $(3.14)(0.0102)^2 = 3.26 \times 10^{-4}$ ft^2; hence,

$$n = \frac{w}{v\rho A_f} = \frac{1.11 \times 10^6}{(3.6 \times 10^3)(50)(3.26 \times 10^{-4})}$$
$$= 1890 \text{ tubes}.$$

Since the total (external) heat-transfer area is 3380 ft^2 and there are 1890 tubes, the total external area per tube is $3380/1890 = 1.78$ ft^2. The external area per ft of tube is $2\pi b$, i.e., $(2)(3.14)(0.0156) = 0.0982$ ft^2; consequently the length of a tube is given by

$$\text{Length of tube} = \frac{1.78}{0.0982} = 18 \text{ ft}.$$

THERMAL STRESSES IN REACTOR COMPONENTS

Introduction

11.191. Thermal stresses are developed in a solid body whenever the expansion or contraction of a differential volume element, that would normally result

from a change in temperature, is prevented. It is convenient to distinguish two different sets of circumstances under which thermal stresses occur. In the first case, the form of the body and the temperature conditions are such that there would be no stress except for the constraint of external forces. An example of this type might be a long fuel element or pipe with little or no freedom of movement at the ends. In the second case, the form of the body and the temperature conditions are such that stresses are produced in the absence of external constraints because of the incompatibility of the natural expansions or contractions of the different parts of the body. Both of these circumstances frequently arise in reactor systems as a result of nonuniform temperature distributions, e.g., in fuel elements, shields, etc., and of mechanical constraints in assembled components.

11.192. A simple form of the general equation which relates the thermal stress to certain physical properties of the solid body and the temperature changes may be derived in the following manner.* The basic procedure is to determine first the dimensions (or shape) the body would assume if unconstrained, and then to calculate the stress, i.e., the force per unit area, required to restore it to its original shape. Consider a rectangular element of an isotropic, elastic material, and suppose, in the first place, that it is free to move in the y- and z-directions, but is constrained in the x-direction. The element is then heated uniformly, so that expansion tends to occur. If there were no restriction in the x-direction, the dimensional increase per unit length, i.e., the unit elongation, would be $\alpha\Delta t$, where α is the thermal coefficient of linear expansion and Δt is the temperature increase. In this case, the unit elongation in the x-direction, represented by ϵ_x, is given by

$$\epsilon_x = \alpha\Delta t.$$

Consequently, if the material is elastic, the stress, σ_x, required to restore the original x-dimension is obtained from Hooke's law as

$$\sigma_x = -E\epsilon_x = -\alpha E\Delta t,$$

where E is Young's modulus for the material. The stresses in the y- and z-directions are, of course, zero.

11.193. When there are restraints upon more than one coordinate, it is necessary to employ the generalized Hooke's law in order to obtain the required relationships. In Cartesian coordinates these are given by the equations

$$\epsilon_x = \frac{1}{E}[\sigma_x - \nu(\sigma_y + \sigma_z)] + \alpha\Delta t$$

* For detailed treatment of thermal stresses, see S. Timoshenko and J. N. Goodier, "Theory of Elasticity," 2nd ed., McGraw-Hill Book Co., Inc., 1951, Chapter 14; S. Timoshenko, "Strength of Materials," Part II, D. Van Nostrand Co., Inc., 1941, pp. 133, 174, 258; L. M. K. Boelter, *et al.*, *op. cit.*, Chapter XVII.

$$\epsilon_y = \frac{1}{E}[\sigma_y - \nu(\sigma_x + \sigma_z)] + \alpha\Delta t$$

$$\epsilon_z = \frac{1}{E}[\sigma_z - \nu(\sigma_x + \sigma_y)] + \alpha\Delta t,$$

where ν is Poisson's ratio. For constraints in the x- and y-directions, $\epsilon_x = \epsilon_y = 0$, whereas in the unrestrained z-dimension, $\sigma_z = 0$. From these conditions it readily follows that

$$\sigma_x = \sigma_y = -\frac{\alpha E}{1 - \nu}\Delta t.$$

11.194. The foregoing analysis may be extended to include restraints in all three directions, and the results may be summarized in the form

$$\sigma = -\frac{\alpha E}{1 - c\nu}\Delta t,$$

where c is 0, 1, or 2, depending upon whether restraints exist in one, two, or three directions, respectively. More generally, the stress is given for a finite system by an expression of the form

$$\sigma = \frac{\alpha E}{1 - c\nu}F(r, t), \tag{11.194.1}$$

in which $F(r, t)$ is a function of the geometry of the body and the temperature distribution within it; examples of some cases of interest are presented below.

11.195. Apart from the temperature and geometrical aspects, it can be seen from equation (11.194.1) that thermal stresses may be kept small by using, as far as possible, materials having low values of the coefficient of thermal expansion, Young's modulus, and Poisson's ratio. Although it is not immediately evident from the general equation, it is reasonable to suppose that a high thermal conductivity is an advantage since this will tend to decrease the temperature differences within the body.

11.196. In the design of reactor components it is frequently desirable to make a preliminary estimate of the order of magnitude of a thermal stress to determine if it is large enough to warrant more precise calculation. For this purpose a relationship similar to equation (11.194.1) may be used; this is

$$\sigma \approx \frac{\alpha E}{1 - c\nu}(\Delta t)_{\max},$$

where $(\Delta t)_{\max}$ is the maximum (numerical) difference between the average temperature and the temperature of any other part of the system.

11.197. The general stress equation given above (and its application to special cases to be described shortly) is satisfactory, provided the material is stressed below the elastic limit throughout. If the elastic limit is slightly exceeded, so that plastic flow occurs, or if creep takes place, the thermal stresses are relieved

in these regions and the values are lower than predicted. In cases where the material undergoes temperature cycling, plastic flow will not be appreciable if the calculated (elastic) stress is less than the sum of the yield strengths in tension and compression. For ductile materials, where these two yield strengths are roughly equal, this means that the calculated stress must not exceed twice the yield strength.

Thermal Stress in Sphere with Uniform Source

11.198. A fairly simple case which is of interest in certain reactor designs is that of a sphere, e.g., of moderator or reflector, in which the heat source is essentially uniform. If a is the radius of the sphere, the temperature t at any point at a radial distance r from the center is given by*

$$t - t_a = \frac{Q}{6k}(a^2 - r^2),$$

where Q is the source strength, k is the thermal conductivity, and t_a is the temperature at the surface of the sphere. If this equation is inserted into the appropriate forms of equation (11.194.1) for a solid sphere† and the required integrations performed, it is found that

$$\sigma_{rad} = \frac{\alpha EQ}{15k(1-\nu)}(r^2 - a^2)$$

and

$$\sigma_{tan} = \frac{\alpha EQ}{15k(1-\nu)}(2r^2 - a^2),$$

where σ_{rad} and σ_{tan} refer to the thermal stresses in radial and tangential directions, respectively. It will be observed that the radial stress becomes zero at the outer surface of the sphere, i.e., when $r = a$, as is to be expected. The tangential stress is zero at a point within the sphere where $2r^2 = a^2$, i.e., where $r = 0.707a$. As an aid to the use of the foregoing equations, the dimensionless quantity $15k(1-\nu)\sigma/\alpha EQa^2$ has been plotted in Fig. 11.24 as a function of r/a for both σ_{rad} and σ_{tan}.‡

Example Calculate the tangential stresses (a) at the surface and (b) at a radial distance of a quarter of the radius, in a 3-in. diameter sphere of beryllium oxide in which heat is generated uniformly at the rate of 10 watts/cm³ due to gamma-ray absorption. The following values of the physical constants of beryllium oxide may be taken as applicable at the existing temperatures:

$\alpha = 5.33 \times 10^{-6}$/°F $E = 39 \times 10^6$ psi

$k = 21.5$ Btu/(hr)(ft²)(°F/ft) $\nu = 0.34$

(a) At the surface of the sphere, $r = a$, so that

$$\sigma_{tan} = \frac{\alpha EQa^2}{15k(1-\nu)},$$

* See Problem 11.6.

† For the derivation of these equations, see S. Timoshenko and J. N. Goodier, *op. cit.*, page 418.

‡ J. C. Carter, Report ANL-4600. This and other figures referred to below are based on unpublished equations derived by R. B. Briggs.

and since a is 1.5 in. = 0.125 ft, and Q is 10 watts/cm³ = 9.66×10^5 Btu/(hr)(ft³), it follows that

$$\sigma_{tan} = \frac{(5.33 \times 10^{-6})(39 \times 10^6)(9.66 \times 10^5)(0.125)^2}{(15)(21.5)(1 - 0.34)}$$

$$= 14{,}700 \text{ psi}.$$

(b) For $r = 0.25a$, i.e., when $r/a = 0.25$, the ordinate for the σ_{tan} curve is found from Fig. 11.24 to be close to -0.86; that is,

$$\frac{15k(1-\nu)\sigma_{tan}}{\alpha EQa^2} = -0.86$$

so that

$$\sigma_{tan} = \frac{-0.86\alpha EQa^2}{15k(1-\nu)}$$

$$= -12{,}600 \text{ psi}.$$

Thermal Stress in Hollow Cylinder with Uniform Heat Source

11.199. An internally cooled fuel element or a moderator piece with regularly spaced cooling holes, as described in § 11.92, may often be treated as an unrestrained hollow cylinder, insulated at the outside, with heat being withdrawn from the inner surface. The radial temperature distribution for this geometry,

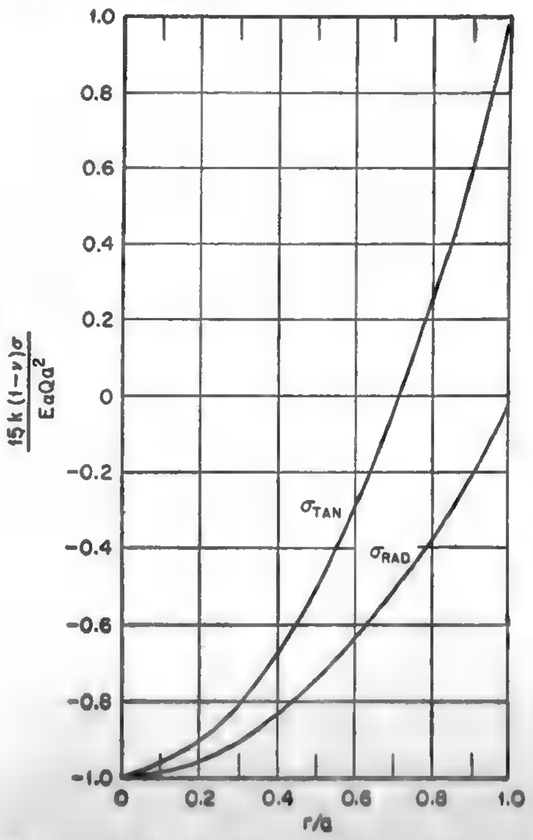


Fig. 11.24 Calculation of thermal stress in solid sphere (J. C. Carter)

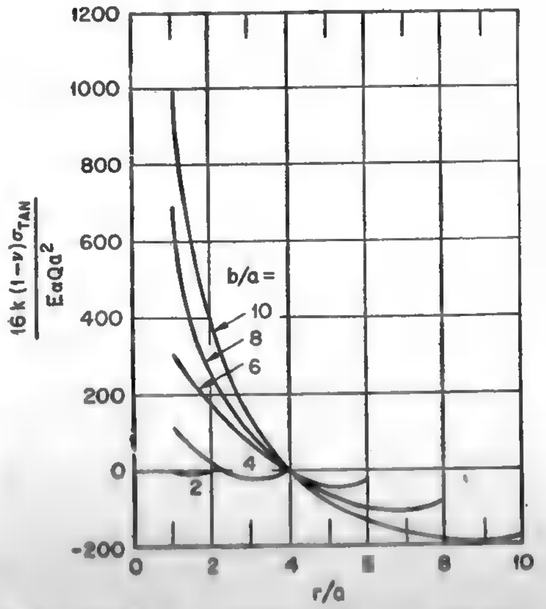


Fig. 11.25 Calculation of tangential thermal stress in hollow cylinder (J. C. Carter)

assuming a uniform source, is given by equation (11.93.1). The results of inserting this into appropriate forms of equation (11.194.1) are shown in Figs. 11.25, 11.26, and 11.27, for a hollow cylinder for which a and b are the internal and external radii, respectively. In these figures the dimensionless quantity

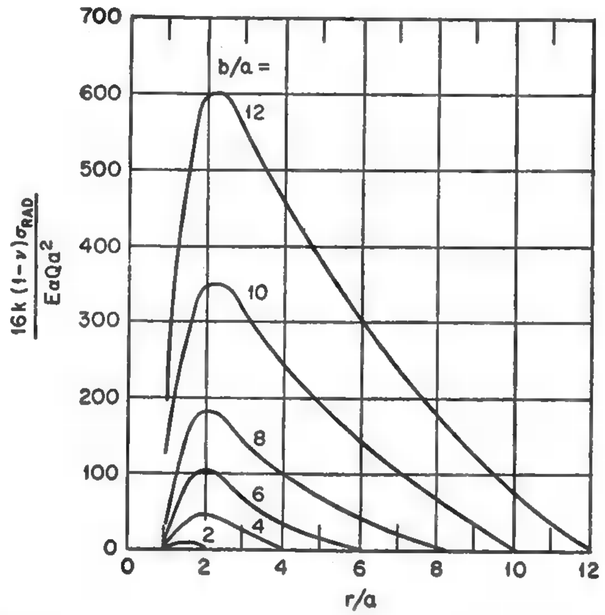


FIG. 11.26 Calculation of radial thermal stress in hollow cylinder (J. C. Carter)

$16k(1-\nu)\sigma/\alpha EQa^2$, for tangential, radial, and longitudinal stresses, is plotted as a function of r/a for various values of the parameter b/a. General inspection of the curves shows that for a given value of r/a, the stresses all increase as b/a, the ratio of the external to the internal radius, increases.

THERMAL STRESS IN SLAB WITH EXPONENTIAL HEAT SOURCE

11.200. The prediction of stresses in a slab heated by an exponentially distributed source is of special interest in connection with the design of thermal shields and of thick-walled pressure vessels. The temperature distribution for a slab of thickness L is given by equation (11.96.1) and combination with the appropriate thermal stress equations* gives

$$\sigma_y \doteq \sigma_z = \frac{\alpha E}{1-\nu} \cdot \frac{Q_0}{k\mu_e^2}\left\{\frac{2x}{L}\left[\frac{3(e^{-\mu_e L}+1)}{\mu_e L} + \frac{6(e^{-\mu_e L}-1)}{(\mu_e L)^2}\right] - \frac{2(e^{-\mu_e L}+2)}{\mu_e L} - \frac{6(e^{-\mu_e L}+1)}{(\mu_e L)^2} + e^{-\mu_e x}\right\}.$$

* S. Timoshenko and J. N. Goodier, *op. cit.*, page 401.

THERMAL STRESS IN UNRESTRICTED CLAD FLAT-PLATE FUEL ELEMENT

11.201. For flat-plate (sandwich type) fuel elements clad on each side (cf. § 11.81), in which the thermal and mechanical properties of the fuel and cladding do not differ greatly, the thermal stress in the y- (and z-) direction (see Fig. 11.28), due to a uniform heat generation in the fuel, is given at the *central plane* of the fuel region by*

$$\sigma_{y\,(\text{at } x=0)} = \sigma_z = -\frac{\alpha EQa^2}{2k(1-\nu)}\left[\frac{c}{a} + \frac{2a}{3(a+c)}\right] \qquad (11.201.1)$$

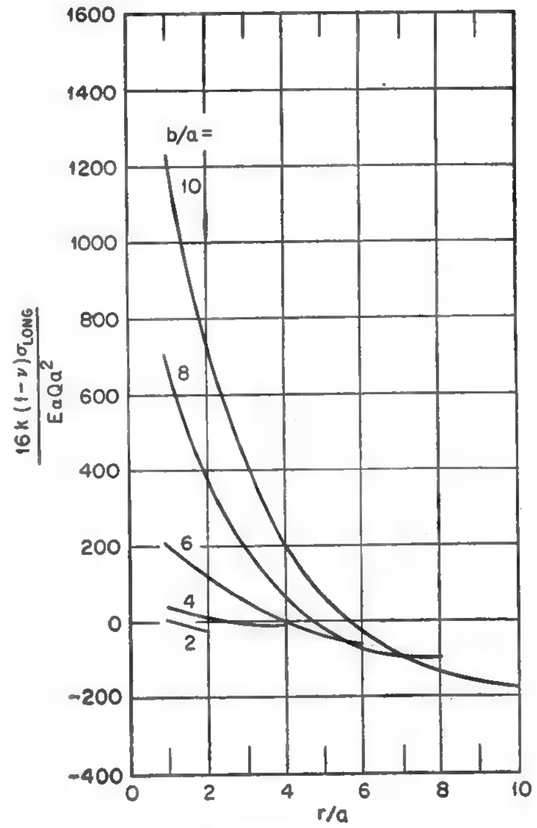


FIG. 11.27 Calculation of longitudinal thermal stress in hollow cylinder (J. C. Carter)

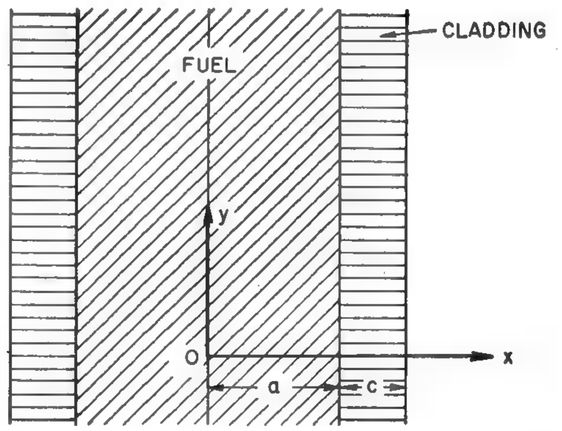


FIG. 11.28 Thermal stress in clad fuel plate

and at the *outer surface* of the cladding by

$$\sigma_{y\,(\text{at } x=a+c)} = \sigma_z = \frac{\alpha EQa^2}{2k(1-\nu)}\left[1 + \frac{c}{a} - \frac{2a}{3(a+c)^2}\right], \qquad (11.201.2)$$

where a is the half-thickness of the fuel region and c is the cladding thickness. (It may be noted that c is equivalent to $b-a$ of Fig. 11.6.)

* P. C. Zmola, unpublished.

Example Calculate the thermal stress (a) at the central plane and (b) at the outer surface of a sandwich-type fuel element consisting of a 0.06-in. thick fuel region clad on both sides with a 0.03-in. layer of a metal having essentially the same mechanical and thermal properties as the fuel region. These properties are as follows:

$$\alpha = 9 \times 10^{-6}/°\text{F} \qquad E = 10 \times 10^{6} \text{ psi}$$
$$k = 10 \text{ Btu/(hr)(ft}^2\text{)(°F/ft)} \qquad \nu = 0.30.$$

Assume that the heat is generated uniformly so as to give a heat flux of 0.5×10^6 Btu/(hr)(ft^2) at the surface of the plate, which is cooled equally on both sides.

It follows from the statement in § 11.83 that the heat flux, i.e., q/A, is equal to aQ, so that in the present case, since $2a$ is 0.06 in., i.e., $a = 2.5 \times 10^{-3}$ ft,

$$Q = \frac{0.5 \times 10^6}{2.5 \times 10^{-3}} = 2 \times 10^{10} \text{ Btu/(hr)(ft}^3\text{)}$$

(a) *Central plane:* From equation (11.201.1),

$$\sigma_y = \sigma_z = -\frac{(9 \times 10^{-6})(10 \times 10^6)(2 \times 10^{10})(2.5 \times 10^{-3})^2}{(2)(10)(1 - 0.3)}\left[\frac{0.03}{0.03} + \frac{2(0.03)}{(3)(0.06)}\right]$$
$$= -9400 \text{ psi.}$$

(b) *Outer surface:* From equation (11.201.2)

$$\sigma_y = \sigma_z = \frac{(9 \times 10^{-6})(10 \times 10^6)(2 \times 10^{10})(2.5 \times 10^{-3})^2}{(2)(10)(1 - 0.3)}\left[1 + \frac{0.03}{0.03} - \frac{2(0.03)}{(3)(0.06)^2}\right]$$
$$= 14{,}700 \text{ psi.}$$

Comparison of Materials

11.202. In concluding this section on thermal stresses it is of interest to compare the expected behavior of different materials in this connection. An examination of the various equations given above shows that in all cases the factor $\alpha E/k(1 - \nu)$ appears, and the value of this quantity for different substances should give a relative indication of the thermal stresses that would be produced under equivalent conditions. Some data for a temperature of about 600°F, together with the approximate tensile strengths at this temperature, are given in Table 11.4. A high value of the thermal stress can be compensated, to some extent, by a large tensile strength.

TABLE 11.4. RELATIVE THERMAL STRESS FACTORS AND TENSILE STRENGTHS

Material	$\alpha E/k(1 - \nu)$	*Tensile Strength* (psi)
Aluminum (2S)	1.4	3,500
Beryllium (extruded)	6.0	41,000
Beryllium oxide	14.6	15,000
Graphite	0.1	3,200
Stainless steel (Type 347)	33.8	69,000
Uranium	24.8	29,000
22% U-Al alloy	2.0	8,100
Zirconium	4.6	15,000

11.203. It is seen from the table that, for a given temperature difference in the material, the corresponding thermal stress will be lowest for graphite, aluminum, and the uranium-aluminum alloy; zirconium and beryllium are in the medium range, whereas beryllium oxide, uranium, and stainless steel (Type 347) will exhibit the largest stresses. It should be noted, however, that the thermal stresses are calculated on the assumption that the material is stretched below the yield point. The ductility of the metal, which permits it to yield ("creep") without cracking, is thus an important consideration, e.g., for stainless steel. Beryllium, on the other hand, is very brittle and cracks if the yield point is exceeded. It is seen, therefore, that the calculated thermal stresses alone do not provide a complete guide to the expected behavior. Although the values are important in establishing temperature conditions under which cracking may occur, they do not indicate whether this actually happens, since other factors are equally significant. Temperature cycling, for example, even below the elastic limit, causes difficulty in many cases, although the thermal stresses may not be excessive.*

Symbols Used in Chapter XI

A	area
A_f	coolant flow area (cross-sectional area of coolant channels)
A_h	heat-transfer area
a	internal radius of hollow tube or cylinder
a	radius of sphere
a	half-thickness of fuel region
b	external radius of hollow tube or cylinder
b	half-thickness of fuel element
C_1, C_2	arbitrary constants
c	factor (in boiling heat transfer)
c	cladding thickness
c_p	specific heat (at constant pressure)
D	pipe diameter
D_e	equivalent (or hydraulic) diameter
E	Young's modulus
E	energy per fission
E_c	energy liberated in capture processes
E_f	energy liberated in fuel per fission
F	shearing force
f	Fanning friction factor
Gr	Grashof number ($= L^3\rho^2 g\beta\Delta t/\mu^2$)
g	acceleration of gravity (or gravitational constant)
H	velocity head

* For further discussion of this subject, see M. Thielsch, "Thermal Fatigue and Thermal Shock," Welding Research Council Bulletin No. 10, April 1952; and articles in "Heat Transfer Symposium," Ann Arbor, Michigan, 1952.

H	height of cylindrical reactor core
H'	effective height of cylindrical reactor
h	heat-transfer coefficient
h_b	boiling heat-transfer coefficient
J_0	Bessel function of zero order
J_1	Bessel function of first order
K_c	pressure loss coefficient in contraction
K_e	pressure loss coefficient in expansion
K_l	pressure loss coefficient in fittings, etc.
k	thermal conductivity
L	length of heat-flow path
L	slab thickness
N	number of nuclei per cm^3
n	number of coolant channels
n	number of heat-exchanger tubes
Nu	Nusselt number ($= hD/k$)
P	power density
P_0	power density at center of reactor
P_{av}	average power density
P_{max}	maximum power density
p	passage perimeter
Δp_f	pressure drop due to friction
Pe	Peclet number ($=$ Re $\times$ Pr $= Dv\rho c_p/k$)
Pr	Prandtl number ($= c_p\mu/k$)
Q	volumetric heat source strength (rate of heat release per unit volume)
Q_0	volumetric heat source at center of reactor (or channel)
q	rate of heat flow
q/A	heat flux
R	thermal resistance
R	radius of cylindrical reactor
R'	effective radius of reactor
r	radial distance (coordinate)
r_0	pipe radius for circulating fuel
r_i	inner radius of annular channel
r_o	outer radius of annular channel
Re	Reynolds number ($= Dv\rho/\mu$)
t	temperature
t_f	mixed-mean fluid temperature
t_s	temperature at solid surface
t_{sat}	saturation temperature of boiling liquid
t_w	temperature at wall of pipe
Δt	temperature difference
Δt_b	surface temperature difference in boiling system

Δt_c	increase in mixed-mean temperature of coolant
Δt_w	temperature difference across wall
U	over-all heat-transfer coefficient
u	fluid velocity (local)
V_h	volume of heat-transfer channel
v	mean velocity of fluid
w	mass (or weight) rate of fluid flow
x, y, z	coordinates
α	fluid thermal diffusivity
α	thermal coefficient of linear expansion
α	function equal to $\pi x/L$
β	thermal coefficient of volume expansion
ϵ	eddy diffusivity
ϵ	roughness factor
ϵ	unit elongation under stress
ϵ_x	unit elongation in x-direction
μ	viscosity
μ_e	energy-absorption coefficient for gamma rays
μ_w	viscosity at wall
ν	Poisson's ratio
ρ	density
ρc_p	volumetric heat capacity
Σ_f	macroscopic fission cross section
σ_f	microscopic fission cross section
σ	thermal stress
$\sigma_x, \sigma_y, \sigma_z$	thermal stress in x-, y-, z-direction
σ_{long}	longitudinal thermal stress
σ_{rad}	radial thermal stress
σ_{tan}	tangential thermal stress
ϕ	neutron flux
ϕ_0	neutron flux at center of reactor
ϕ_{av}	average neutron flux
ϕ_{long}	longitudinal neutron flux
∇^2	Laplacian operator

PROBLEMS*

1. In a heat exchanger consisting of steel tubes, with $k = 25$ Btu/(hr)(ft^2)(°F/ft), having an internal diameter of 1.1 in. and external diameter 1.3 in., heat is transmitted from a primary coolant, inside the tubes, to a secondary coolant, outside. The respective heat-transfer coefficients are 8400 and 6300 Btu/(hr)(ft^2)(°F). Determine the temperature difference between the primary and secondary coolants and the temperature drop across the tube wall when heat is being transferred at a rate of 5000 Btu/hr per ft of tube.

* If not supplied, data required for the solution of the problems will be found in the text or in the tables in the Appendix.

2. A cylindrical, 1-in. diameter, fuel element of natural uranium is clad with aluminum 0.05 in. thick and is cooled with water. The temperature of the water at the surface of the element is 155°F, the heat-transfer coefficient is 6600 Btu/(hr)(ft²)(°F), and the thermal flux in the reactor is 10^{14} neutrons/(cm²)(sec). Assuming the thermal source in the fuel to be uniform, determine the temperature distribution through the fuel and cladding.

3. If the cooling water in the preceding problem flows through an annular channel surrounding the fuel element at a rate of 20 ft/sec, estimate the outside diameter of the annulus which will make the heat-transfer coefficient equal to the value given.

4. A plate-type fuel element, consisting of a uranium-zirconium alloy 3 mm thick clad on each side with a 0.5 mm thick layer or zirconium, is cooled by water (under pressure) at 400°F, the heat-transfer coefficient being 7500 Btu/(hr)(ft²)(°F). If the temperature at the center of the fuel must not exceed 1050°F, determine the maximum specific power attainable, expressed in kw/kg of uranium-235, given that the fuel region contains 10 per cent by weight of the latter. Determine the temperature distribution through the fuel and cladding when the reactor is operating at the maximum power. (The density of the zirconium may be taken as 6.5 g/cm³ and its thermal conductivity as 12 Btu/(hr)(ft²)(°F/ft); the latter value may be taken to apply also to the fuel region.)

5. In the preceding problem the resistance at the fuel-cladding interface is neglected. Investigate the effect of a possible surface conductance of 10,000 Btu/(hr)(ft²)(°F).

6. Show that for a sphere of radius a and thermal conductivity k having a uniform volumetric heat source Q, the temperature, t, at a distance r from the center, is given by

$$t - t_a = \frac{Q}{6k}(a^2 - r^2),$$

where t_a is the temperature at the exterior.

7. A beryllium moderator ($k = 7.2$ Btu/(hr)(ft²)(°F/ft)) is cooled by water at 100°F flowing through tubular channels of 1-in. diameter at the rate of 15 ft/sec. If the maximum temperature in the moderator is 180°F, what is the rate, expressed in kw/ft³, at which heat is removed from the beryllium? The moderator may be treated as equivalent to an insulated cylinder of 1 ft. diameter with a cooling channel through the center.

8. Gamma rays, with an energy flux of 2×10^{13} Mev/(cm²)(sec), are incident upon an iron thermal shield 4 in. thick. Cooling air maintains the temperature at the face nearest the reactor at 150°F, whereas that at the other face is 100°F. Taking the energy absorption coefficient of iron for the given gamma rays to be 0.16 cm^{-1} and the thermal conductivity to be 30 Btu/(hr)(ft²)(°F/ft), determine the location and value of the maximum temperature in the thermal shield.

9. Using the data in § 11.101, estimate the temperature drop at the interface between a fuel slug and the graphite in the Oak Ridge natural uranium-graphite reactor when operating at a power of 3800 kw. It may be assumed that 1000 fuel channels are filled to an average length of 14 ft.

10. A reactor cooled with liquid sodium is to produce 200,000 kw of heat. The cylindrical fuel elements are 8 ft long and 0.75 in. in diameter, and the coolant flows through an annular channel of 0.625-in. outer radius at a rate of 25 ft/sec. The liquid sodium enters the reactor at 700°F and the maximum permissible surface temperature of the fuel rods is set at 1050°F. Calculate the minimum number of cooling channels that would be required and the average exit temperature of the coolant. (For the purpose of this calculation the reactor core may be treated as a bare cylinder.)

11. Suppose that in the preceding problem the liquid sodium flows into the channels from a large pipe (or header) and flows into a similar pipe when it leaves the reactor. Calculate the pumping power (*a*) in kw, (*b*) in theoretical hp, required to pump the coolant through the reactor.

12. Calculate the values of the heat-transfer coefficient for (*a*) water at 450°F, (*b*) sodium at 800°F, at flow velocities of 10, 20, 30, and 40 ft/sec through a circular tube having an internal diameter of 1 in. What conclusions may be drawn concerning the advantage of increasing the flow velocity, taking into account the required pumping power, for the two cases?

13. Using the method outlined in § 11.163, derive an expression whereby the fraction of reactor heat required for pumping the coolant may be compared for different (nonmetal) coolants on the basis of their known physical properties. In the derivation it is to be assumed that the reactor operates at the same power, that the length, diameter, and number of the coolant passages are the same, as also are the temperature increase of the coolant and the temperature drop between the fuel-element surface and the coolant, for all the coolants. The conditions are to be taken such that turbulent flow occurs in every case.

14. In a reactor of the Swimming Pool type, the fuel plates are 0.25 in. apart and the total flow area for the water is 0.80 ft². The temperature of the water at the bottom of the fuel plates is 85°F. What will be the temperature at the top when the reactor operates at a power level of 100 kw?

15. Calculate the heat-transfer coefficient of helium at 500°F and 200 psia flowing at a rate of 150 ft/sec through a channel of rectangular cross section, 0.2×3.0 in. Consider, taking into account the effect on the pumping power, what changes might be made either in the design of the coolant channels or in the conditions of the coolant that would result in a beneficial change in the heat-transfer coefficient.

Chapter XII *

NUCLEAR REACTOR DESIGN VARIABLES

MAIN ASPECTS OF REACTOR DESIGN

Introduction

12.1. The complete design of a nuclear reactor system is a lengthy and complicated procedure involving the cooperation of many engineers and scientists possessing specialized knowledge and experience of various types. Fundamentally, the operation requires a synthesis of information from the various fields which have been outlined in the preceding chapters of this book. If circumstances permit, due regard should be paid to economic considerations such as are characteristic of engineering practice in general. Good reactor design should take into account the cost and availability of materials, the fabrication of fuel elements and their processing after discharge from the reactor, the amount of fuel held up during various stages, and the cost of constructing and operating the reactor, in addition to the more obvious problems of criticality, heat removal, controls, shielding, etc.

12.2. It is not possible in this chapter to refer to all the details of reactor design. It is proposed, rather, to examine the main variables concerned and to review the data and arguments upon which reasonable choices can be based. Although many aspects of reactor design are interrelated, it is nevertheless convenient to consider these aspects in three broad categories; they are (1) the nuclear system, (2) the heat-removal system, and (3) structural, mechanical, and associated systems.†

* Reviewed by R. B. Briggs, J. A. Lane, and N. F. Lansing.

† For general reviews of reactor design problems, see W. F. Davidson, *Nucleonics*, **5**, No. 5, 4 (1949); L. A. Ohlinger, *ibid.*, **5**, No. 6, 38 (1949); **6**, No. 1, 10 (1950); **6**, No. 2, 54 (1950); **6**, No. 3, 46 (1950); K. H. Kingdon, *ibid.*, **10**, No. 4, 18 (1952); J. A. Lane and S. McLain, *Chem. Eng. Prog.*, **49**, 287 (1953); S. McLain, *ibid.*, **50**, 240 (1954); W. E. Abbott, *ibid.*, **50**, 245 (1954); F. T. Miles and I. Kaplan, *Chem. Eng. Prog. Symposium Series*, No. 11, **50**, 159 (1954); W. E. Parkins, *ibid.*, No. 13, **50**, 53 (1954); R. L. Witzke and S. A. Haverstick, *Elec. Eng.*, **74**, 116 (1955); S. McLain, *ibid.*, **74**, 44, 144 (1955).

The Nuclear System

12.3. After the purpose of the reactor and its maximum operating power have been established, the initial reactor concept will include, at least, provisional decisions concerning the nature of the principal components, namely, fuel, moderator, reflector, and coolant. These decisions are bound up with the question of the neutron energy range (or spectrum), i.e., fast, intermediate, or slow, in which most of the fissions are to occur. With this information preliminary calculations can be made of the actual masses of fuel for various proportions and arrangements of fuel and moderator, and the prescribed reflector material, as outlined in Chapter III. These estimates will be refined as the design proceeds and information becomes available concerning the thermal aspects of the system, e.g., nature of the coolant, heat-exchanger area, design of fuel elements, etc.

Heat-removal System

12.4. A decision must first be made with regard to the general method of heat removal, e.g., by internal cooling of the reactor core or by circulating the fuel and cooling it externally. If an internal coolant is to be used, this must be chosen, as well as the heat-transfer mechanism, e.g., natural convection, forced convection, or boiling. The arrangement of the fuel and moderator and the fuel-element design are selected in order to facilitate the transfer of heat to the coolant. As indicated in Chapter XI, the requirements here may be in conflict with those based on optimum nuclear conditions, so that some compromise may prove necessary.

12.5. The operating temperatures of the reactor, i.e., the inlet and outlet temperatures of the coolant, will be determined by the rate at which the coolant flows through the reactor core, by the physical properties of the coolant, and by the temperature of the medium to which heat is transferred in the external heat-exchange system. The coolant outlet temperature is usually limited by the maximum permissible temperature of the fuel elements (or other solid components) in heterogeneous reactors, or by the pressure of the system, by chemical stability, or by corrosion considerations in homogeneous reactors. Since high temperatures are desirable in a power reactor, for thermodynamic reasons, the coolant temperatures are maintained as high as possible subject to the foregoing limitations.

Structural, Mechanical, and Associated Systems

12.6. The choice of the fuel cladding, which is part of fuel-element design, is also, in a sense, one of the structural problems. A solid moderator, too, is part of the over-all reactor structure. Among other aspects of structural and mechanical problems are the design of the instrumentation and control systems, the shield, the pressure vessel (if required), the heat exchanger (or boiler), and methods for loading and unloading the reactor fuel. Since instrumentation,

control, and shielding have been treated in preceding chapters, they will not be considered further here. Conventional engineering topics, i.e., problems of design and construction not directly influenced by radiation, will also be omitted. The discussion will, therefore, be restricted largely to the variables involved in the design of the reactor core.

REACTOR EXPERIMENTS

Purpose of Reactor

12.7. Reactors which have already been constructed or have been given serious consideration fall into five general groups according to the purpose for which they are intended. Although there may be some overlap in functions, these may be described as (1) reactor experiments, (2) experimental (or research) reactors, (3) production reactors (or converters), (4) reactors for power only (mobile and central station), and (5) dual-purpose (power and production) reactors. The design problems to be treated in this chapter will refer more particularly to power reactors. However, for the sake of completeness, a general review of the factors determining the design of reactor experiments and experimental (or research) reactors will be presented first. More detailed descriptions of these and other reactors will be given in Chapter XIII.

12.8. The purpose of a reactor experiment is to obtain information concerning a specific design or concept. The first self-sustaining nuclear reactor to be constructed, namely, CP-1 (Chicago Pile No. 1) was a reactor experiment; its objective was to confirm the expectation that a fission chain reaction could be maintained in a natural uranium-graphite system. More recently, two important reactor experiments have been the EBR (Experimental Breeder Reactor), to test the possibility of simultaneous breeding and power production, and the HRE (Homogeneous Reactor Experiment), to provide information concerning the operation and nuclear stability of an aqueous circulating-fuel reactor to be used as a source of power.

Reactor Development Program

12.9. The Five-Year Power Reactor Development Program, proposed by the U. S. Atomic Energy Commission in 1954, involves a number of reactor experiments. These include EBR-2 and HRE-2 (or Homogeneous Reactor Test, HRT), which are larger-scale versions of the experiments mentioned in the preceding paragraph. A further development of the latter experiment will be a study of thermal breeding of uranium-233 from thorium. In addition there will be an experimental boiling water reactor (EBWR) which will explore further (cf. § 8.238) the prospect of obtaining steam by boiling water directly within the reactor, and a sodium-graphite experiment (SRE) to investigate high-

temperature operation of a reactor using graphite as moderator and sodium as coolant.*

RESEARCH REACTORS

Requirements of Research Reactors

12.10. An experimental or research reactor is essentially a laboratory device; it can be used to provide physicists and engineers with some basic experience in reactor design and operation, and also to perform a variety of experiments and investigations requiring relatively strong neutron sources.† After its reconstruction as CP-2, early in 1943, at the Argonne National Laboratory, the first reactor experiment (CP-1) became an experimental and research reactor and was extensively used to determine the nuclear properties of various possible reactor materials and for early studies of shielding, controls, instrumentation, etc. The requirements of a research reactor are essentially safety, simplicity, and reliability in operation, accessibility for experiments, a relatively high neutron flux, and moderate cost. Reasonably small size, if compatible with accessibility, is also desirable.

12.11. According to equation (2.164.1), the flux, ϕ, of the neutrons causing fission in a reactor is related approximately to the power (P watts) and the mass (g grams) of fissionable material, e.g., uranium-235, by

$$\phi = \frac{P}{8.3 \times 10^{10} g\sigma_f} = \frac{1.2 \times 10^{13} P}{g\sigma_f(\text{barns})}, \tag{12.11.1}$$

where σ_f is the value of the microscopic fission cross section expressed in cm²/nucleus and σ_f(barns) is the value in barns. In order to minimize the power which must be removed from the reactor at a given flux, it is desirable to keep w, the mass of fissionable material in the core, as small as possible. This can be achieved by increasing the enrichment of uranium-235 in the fuel to the point where neutron absorption in uranium-238 is relatively unimportant (20 per cent uranium-235 or more) and using a good moderator. As long as natural uranium as fuel and graphite as moderator were the only materials available for reactor construction, the design of experimental and research reactors was restricted. With the availability of heavy water and highly enriched uranium, the situation has changed, as will be seen in the following paragraphs.

12.12. From the design standpoint, most research reactors constructed hitherto fall into five main classes: (1) natural uranium-graphite, (2) natural

* "Report on the Five-Year Power Reactor Development Program Proposed by the Atomic Energy Commission," U. S. Government Printing Office, March 1954.

† For possible uses of experimental reactors, see D. J. Hughes, "Pile Neutron Physics," Addison-Wesley Press, Inc., 1953; C. Beck, *Nucleonics*, **9**, No. 5, 18 (1951); M. Fox, *ibid.*, **11**, No. 6, 46 (1953); L. D. P. King, Report AECU-2000, p. 81; R. L. Murray, *ibid.*, p. 217. Several other papers in Report AECU-2900 are also of interest in this connection. See also, M. Benedict, *Chem. Eng. Prog.*, **51**, 53F (1955).

uranium-heavy water, (3) enriched uranium-heavy water, (4) homogeneous enriched uranium-ordinary water (Water Boiler), and (5) heterogeneous enriched uranium-ordinary water (MTR and Swimming Pool). The general characteristics of these five types will be outlined below. Reference will also be made to a few other reactor designs which do not fall into these categories.

Natural Uranium-Graphite Type

12.13. In addition to CP-2, the Oak Ridge X-10 and Brookhaven National Laboratory (BNL) reactors in the United States use natural uranium as fuel and graphite as moderator. The Graphite Low-Energy Experimental Pile (GLEEP) and the British Experimental Pile (BEPO), in England, also employ the same materials. In each case, with the exception of CP-2, cooling is by forced convection of air. The normal operating power of CP-2 is very low (2 kw) because it has no provision for heat removal except conduction through the graphite moderator and the concrete shield.

12.14. The main advantage of the air-cooled, natural uranium-graphite type of experimental reactor lies in its large size and adaptability, so that many observations of different types can be performed simultaneously. Large pieces of apparatus can be readily accommodated alongside the reactor, and even included within the core, if necessary, by removing some of the graphite. The X-10 and BNL reactors have also been used extensively for the large-scale production of radioisotopes, by neutron reactions with a variety of elements. On the other hand, the large size and weight represent the chief drawbacks of reactors of this type. The minimum amounts required for criticality are about 30 tons of natural uranium metal and 350 tons of graphite, including the reflector, although the actual loadings are roughly twice as large. The over-all size of the BNL reactor, for example, with its shield, is $38 \times 55 \times 30$ ft, and its total weight, with the concrete foundations, is about 20,000 tons.

Natural Uranium-Heavy Water Type

12.15. Since heavy water is a better moderator than graphite and also has a smaller cross section for neutron absorption, the infinite multiplication factor of a natural uranium-heavy water system is appreciably greater than one consisting of natural uranium and graphite (see Chapter III). Consequently reactors with heavy water as moderator are considerably smaller than those employing graphite in this capacity. Thus, the first heavy water reactor to be constructed, CP-3 (Argonne National Laboratory), contained 3 tons of natural uranium metal and 6.5 tons (1500 gal) of heavy water in an aluminum tank 6 ft in diameter and nearly 9 ft high. Because of the smaller mass of fissionable material, the average thermal neutron flux in CP-3, operating at 300 kw, was about the same as in the moderator of the X-10 reactor at 3800 kw, i.e., 5×10^{11} neutrons/(cm^2)(sec).

12.16. For various reasons, research reactors with heavy water have proved

attractive. They provide a relatively high neutron flux for a given power, since they require less uranium metal than do graphite reactors. This latter consideration is important in countries where heavy water is more readily available than uranium. The Canadian ZEEP (Zero Energy Experimental Pile) and the NRX (National Research Experimental) Reactor both employ natural uranium as fuel and heavy water as moderator. The latter (NRX) is somewhat unusual in the respect that ordinary water is the coolant, flowing through annular spaces around the fuel rods in the reactor. In most other heavy-water reactors the fuel elements are cooled by convection to the moderator, which is circulated and cooled in an external heat exchanger. An exception is the French Saclay (P-2) reactor, referred to below, in which a gas is the coolant within the reactor. In the internally cooled NRX reactor, the heavy water moderator is also circulated and cooled externally.

12.17. Among other research reactors of the natural uranium-heavy water type, mention may be made of the French ZOE (Chatillon) and the gas-cooled P-2 (Saclay) reactors, the Joint (Norwegian-Dutch) Establishment for Nuclear Energy (JEEP) reactor at Kjeller, Norway, and the Swedish reactor at Stockholm. The JEEP and Swedish reactors are similar in many respects to CP-3.

Enriched Uranium-Heavy Water Type

12.18. When the CP-3 reactor was dismantled in 1950, because of suspected corrosion of the aluminum canning of the fuel rods, the natural uranium was replaced with metal (2% U-98% Al) enriched in uranium-235. The resulting system, referred to as CP-3′, was thus the first reactor to use enriched fuel in conjunction with heavy water as moderator. The enrichment of the fuel made possible a decrease in the total weight of uranium metal and a four-fold increase in the average thermal-neutron flux, to 2×10^{12} neutrons/(cm^2)(sec), for the same power output (300 kw).

12.19. As a replacement for CP-3′, Argonne National Laboratory has built a reactor of somewhat similar type, called CP-5, with heavy water as moderator, but using plate-type fuel elements of highly enriched uranium metal alloyed with aluminum. This design was chosen chiefly because of the good performance and excellent characteristics for research of the CP-3′ reactor. As a consequence of the high enrichment of the fuel, the core of CP-5, which is approximately cylindrical, is quite small; it is 2 ft in height and the same in diameter. The reactor contains only 1.2 kg of uranium-235 and 6.5 tons of heavy water (including a 2-ft thick reflector) and, when operating at its normal power of 1000 kw, the average thermal-neutron flux is 2×10^{13} neutrons/(cm^2)(sec).

Homogeneous Enriched Uranium-Ordinary Water (Water Boiler) Type

12.20. The original so-called Water Boiler was put into operation at Los Alamos Scientific Laboratory in 1944. The core contained about 13 liters

(3½ gal) of a solution in ordinary water of about 6 kg of uranyl sulfate enriched in uranium-235 to the extent of 14.6 per cent (0.57 kg of U-235); the container was a stainless steel sphere about 1 ft in diameter. The name Water Boiler has been applied to reactors of this type, but it is misleading since the temperature of the water, which is above normal because of the heat generated by fission, is maintained below the boiling point, in the interest of stability of operation. The Los Alamos Water Boiler was the first reactor to use enriched uranium as fuel, as well as the first to employ ordinary water as moderator. As stated in § 3.184, it is apparently not possible to achieve a critical system with natural uranium and ordinary water, so that enrichment of the fuel is necessary.

12.21. The original (LOPO) Water Boiler had no cooling system and operated at very low power (<1 watt). Upon its reconstruction as HYPO later in 1944, the fuel solution contained somewhat more than 10 kg of uranyl nitrate (0.87 kg U-235) in about 13 liters, and cooling coils through which water could be circulated were introduced. The normal operating power was then raised to 6 kw. In a further improvement, known as SUPO, made in 1951, uranium of about 90 per cent enrichment was used, in the form of roughly 1.5 kg of uranyl nitrate (0.87 kg U-235). The operating power is 45 kw, and the average thermal-neutron flux is 10^{12} neutrons/(cm^2)(sec).

12.22. The simple design of the Water Boiler, with a core consisting merely of an aqueous solution, and its intrinsic safety, due to its large negative temperature coefficient, have made this reactor of special interest as an experimental device for universities and industrial laboratories. The first such reactor to be built was at North Carolina State College, Raleigh, N. C., and it commenced operation in 1953. It was originally called the North Carolina State Reactor (NCSR), but is now known as the Raleigh Research Reactor (RRR). It employs 0.86 kg of uranium-235, in the form of uranyl sulfate (93 per cent enrichment), in about 13 liters of aqueous solution, contained in a stainless steel cylinder. With the aid of four cooling coils in parallel, the normal operating level is maintained at about 10 kw; the average thermal flux is then about 2×10^{11} neutrons/(cm^2)(sec).

12.23. A compact model of the Water Boiler reactor, called a Water Boiler Neutron Source (WBNS), is available commercially. It is intended for use in research laboratories where especially high neutron fluxes are not required. The average flux in regular operation is 2×10^7 thermal neutrons/(cm^2)(sec) and the power is about 1 watt.

Heterogeneous Enriched Uranium-Ordinary Water Type

12.24. The Materials Testing Reactor (MTR) was the first reactor designed to employ solid enriched uranium fuel elements with ordinary water as moderator, but, as will be seen below, it was not the first to go into operation. The MTR, completed in 1952, was intended primarily for studying the effects of nuclear radiations on materials required for reactor construction. It was designed, therefore, to provide a higher neutron flux than any existing reactor. The fuel elements of the MTR consist of thin curved plates of an alloy of highly enriched uranium (~93 per cent uranium-235) with aluminum, clad on each side with aluminum to prevent escape of fission products and to protect the alloy from corrosion. The fuel plates are arranged parallel to one another, with the spaces between serving as channels for water which acts as both moderator and coolant. The core of the MTR contains only about 4 kg of uranium-235, but with forced convection cooling the reactor operates at 30,000 kw; the average thermal flux is then 2×10^{14} neutrons/(cm^2)(sec).

12.25. Prior to the completion of the MTR, a mock-up had been built at Oak Ridge National Laboratory in 1950, to test various features of the MTR design. This operated so successfully that it was retained, temporarily at least, with some modification, as the Low Intensity Test Reactor (LITR), operating at 3000 kw. Soon afterward, when a new shield testing facility was required at Oak Ridge National Laboratory, it was decided to use a reactor core, similar to that of the LITR, as the radiation source. This core is suspended in a large tank of water ($20 \times 40 \times 20$ ft deep), which is the moderator as well as the free-convection cooling medium. Because of this configuration, the reactor, which was completed early in 1951 and originally referred to as the Bulk Shielding Reactor (BSR), is now often called the Swimming Pool Reactor. The same name has been applied to reactors of similar construction but having fuel plates of a somewhat different design. The core of the reactor is only about $1 \times 1\frac{1}{2} \times 2$ ft in size and contains 3.2 kg (or less) of uranium-235 as 90 per cent enriched material alloyed with aluminum. The active lattice can be surrounded on its four faces by a beryllium oxide (or other) reflector, if required.

12.26. An important aspect of reactors of the Swimming Pool type is the fact that they can operate at powers up to 1000 kw, at least, using only free convection of the water in the tank (or pool) for cooling purposes; the average flux is then nearly 10^{13} neutrons/(cm^2)(sec). By circulating the water through an external heat exchanger, both power output and flux can be readily increased.* The design of the Swimming Pool reactor core (see § 13.70) is such that considerable flexibility in fuel loading and in the reflector is possible. Although the negative temperature coefficient is not so large as for the Water Boiler reactor, it is nevertheless sufficient to insure safety in operation. Even a sudden moderate increase in reactivity, due to an accident, is not likely to have serious consequences, as has been shown by the Boiling Water Reactor experiments (§ 12.149).

Comparison of Swimming Pool and Water Boiler Reactors

12.27. Like the Water Boiler, the Swimming Pool reactor has attracted much interest as an experimental device for university and research laboratories.

* By making provision for dealing with the induced activity due to nitrogen-16 in the water, an operating power greater than 1000 kw is possible with free-convection cooling.

Since each of these two reactor types has its proponents, some of the advantages and disadvantages of each will be reviewed here.* Because of the instability resulting from the presence of hydrogen and oxygen gas bubbles, due mainly to decomposition of the water by fission fragments, the power level of the Water Boiler is possibly limited, even with forced cooling, to 50 kw and the thermal flux to an average of about 10^{12} neutrons/(cm^2)(sec).† There is also the possibility that, at high powers (and normal temperatures), some of the uranium may be precipitated as peroxide, due to the increase in concentration of hydrogen peroxide (§ 8.141), and thus affect the criticality. No such limitations apply to the Swimming Pool reactor, in which a flux of 10^{12} neutrons/(cm^2)(sec) is common, using only natural convection for cooling. Radiation decomposition of the water is not important, since the fission products are held within the sandwich-plate type fuel elements. This avoids the necessity for a well-shielded recombining system (§ 8.148), such as is required for the Water Boiler.

12.28. One of the advantages claimed for the Swimming Pool reactor is its flexibility. The core can be moved to various locations within the tank, and experimental facilities can be placed in the water surrounding the reactor. Thus, the Swimming Pool reactor lends itself particularly to shielding studies — which was its original purpose — whereas the Water Boiler, in its usual form, does not. However, there would appear to be no serious objections to mounting a Water Boiler in a large tank of water, instead of using the solid moderator, reflector, and shield hitherto employed.

12.29. In certain respects the Water Boiler presents a more severe radiation hazard than does the Swimming Pool reactor; this is because the radioactivity of the fuel solution, due to the accumulation of fission products during operation, is much greater than that induced in the pool water by neutron capture. A leak of the fuel solution from the Water Boiler, although not very probable, could have serious consequences. Similarly, there is a possibility that radioactive gases, e.g., xenon and krypton, might escape from the gas-recombiner system. On the other hand, it is believed that the Water Boiler is less likely to go out of control than a reactor of the Swimming Pool type.

12.30. If the cost of the fuel were a consideration, the advantage would lie with the Water Boiler. This reactor requires less than 1 kg of uranium-235 for satisfactory operation. It is probable that a plate-type system could be made critical with the same amount of uranium-235, but, with present designs, the benefits of the Swimming Pool reactor can be realized only if 2 to 3 kg of uranium-235 are available. Another point in favor of the Water Boiler is that, since the fuel is in solution, there are no fuel elements requiring special fabrication.

* Complete reactor installations of both types are available commercially. Any approved university or research institution can operate a reactor facility, provided certain conditions required by the Atomic Energy Commission can be met. The A.E.C. will then lend the uranium-235 fuel without cost (cf. T. H. Johnson, Report AECU-2000, p. 3).

† In one commercial proposal, a maximum power of 200 to 400 kw is claimed as the result of a cooling system of special design (*Chem. Eng. News*, **32**, 3002 (1954)).

Processing of the spent fuel is also greatly simplified. However, as long as the Atomic Energy Commission is prepared to fabricate and lend fuel elements to approved institutions without charge, these matters are not significant to the operators of the reactor.

Other Research Reactors

12.31. There are a few research reactors which do not belong in any of the main categories considered above. One of these is the (NAA) homogeneous graphite reactor designed by North American Aviation, Inc.* The core consists of graphite blocks through which enriched uranium oxide (U_3O_8) is uniformly dispersed; the coolant is heavy water. The normal operating level is 160 kw and the average thermal flux is 10^{12} neutrons/(cm^2)(sec). At this power the core should last more than 20,000 hours before reloading is necessary. Because of the larger volume of the core, as compared with the Water Boiler and Swimming Pool reactors, more experimental facilities can be provided. One of the prime considerations in the NAA design is safety in operation.

12.32. A very compact and relatively inexpensive reactor, which can be used for neutron absorption measurements (§ 13.106), as well as a neutron source, is the Thermal Test Reactor (TTR) of the Knolls Atomic Power Laboratory. The fissionable material is enriched uranium and several substances, e.g., polyethylene, hydrocarbon oil, and water, act as moderators. The TTR is unusual in the respect that the "core" fits into a cylindrical annular space. An internal reflector provides a valuable experimental medium. The power level of the reactor, which has no special cooling, is 0.1 kw and the *maximum* thermal flux is 3.4×10^9 neutrons/(cm^2)(sec).

12.33. In all the experimental reactors reviewed so far, the fissions are produced mainly by thermal neutrons. The first, and for several years the only, reactor wherein the fission chain was maintained by fast neutrons was the Los Alamos Fast Reactor, which went into operation late in 1946 and was dismantled in 1953. The fuel consisted of relatively pure plutonium rods cooled by liquid mercury; no elements of small mass number, which would slow down the neutrons, were present. The operating power level was 25 kw and the maximum fast flux was about 10^{13} neutrons/(cm^2)(sec). The initial purpose of the Fast Reactor was to provide information relating to fast-neutron properties to be used in weapons development. However, data were obtained which could be applied in the design of other fast-neutron reactors, such as the EBR.

Summary

12.34. For reference purposes the general characteristics of experimental and research reactors, for which data are available, are summarized in Table 12.1.

* A. M. Stelle, *Proc. Conf. on Nucl. Eng.*, Berkeley, 1953, A-15; *Science*, **119**, 15 (1954).

TABLE 12.1. SUMMARY OF EXPERIMENTAL REACTORS*

Natural Uranium-Graphite Type

Designation	Uranium (tons)	Graphite (tons)	Power Level (kw)	Av. Thermal Flux neutrons/(cm²)(sec)
CP-1	40	385	—	—
CP-2	45	472	2	3×10^{8}
X-10	54	620	3800	5×10^{11}
BNL	60	700	28,000	3×10^{12}
GLEEP	33	505	100	3×10^{10}
BEPO	40	~600	6000	10^{12}

Natural Uranium-Heavy Water Type

Designation	Uranium (tons)	Heavy Water (tons)	Power Level (kw)	Av. Thermal Flux neutrons/(cm²)(sec)
CP-3†	3	6.5	300	5×10^{11}
ZEEP	—	—	0.0035	5×10^{6}
NRX	10.5	17	30,000	2×10^{13}
ZOE	~3	~5	5	2×10^{10}
Saclay (P-2)	~3	~6	1000	5×10^{12}
JEEP†	~2.5	7	300	10^{12}

† The Swedish reactor is similar to CP-3 and JEEP in many respects.

Enriched Uranium-Heavy Water Type

Designation	Uranium-235 (pounds)	(kg)	Heavy Water (tons)	Power Level (kw)	Av. Thermal Flux neutrons/(cm²)(sec)
CP-3′	9	4.1	7	300	2×10^{12}
CP-5	2.6	1.2	6.5	1000	2×10^{13}

Water Boiler Type

Designation	Uranium-235 (pounds)	(kg)	Solution Volume (gallons)	Power Level (kw)	Av. Thermal Flux neutrons/(cm²)(sec)
LOPO	1.3	0.59	3½	<0.001	$\sim 10^{6}$
HYPO	1.9	0.87	3½	6	10^{11}
SUPO	1.9	0.87	3½	45	10^{13}
RRR (NCSR)	1.9	0.87	3½	10	2×10^{11}
WBNS	1.9	0.87	3½	0.001	2×10^{7}

* Data obtained mainly from H. S. Isbin, *Nucleonics*, **10**, No. 3, 10 (1952); **11**, No. 6, 65 (1953); J. A. Lane, *ibid.*, **12**, No. 4, 12 (1954); Report AECU-2000, p. 66.

TABLE 12.1. SUMMARY OF EXPERIMENTAL REACTORS (continued)

MTR and Swimming Pool Types

Designation	Uranium-235 (pounds)	(kg)	Core Volume (ft³)	Power Level (kw)	Av. Thermal Flux neutrons/(cm²)(sec)
MTR	9	4.1	~5	30,000	2×10^{14}
LITR	6.5	3	~5	3000	2×10^{13}
BSR‡	6	~3	2–3	100	5×10^{11}

‡ These figures apply generally to reactors of the Swimming Pool type.

Other Thermal Types

Designation	Uranium-235 (pounds)	(kg)	Core Volume (ft³)	Power Level (kw)	Av. Thermal Flux neutrons/(cm²)(sec)
NAA	7	3.2	—	160	$\sim 10^{12}$
TTR	6	2.7	~3.6	0.1	$\sim 10^{9}$

The average thermal fluxes quoted usually refer to values in the moderator or in experimental holes; the thermal fluxes in the fuel are invariably lower. It should be noted that in all thermal reactors there is an appreciable fast-neutron (>0.1 Mev) flux, amounting to one tenth to one half of the thermal flux. The fast flux is always higher in the fuel than in the moderator.

PRODUCTION REACTORS OR CONVERTERS

Purpose of Production Reactors

12.35. In a production reactor, fertile material, e.g., uranium-238, is converted into a fissionable material, e.g., plutonium-239, at the expense of another fissionable species, e.g., uranium-235, without necessarily attempting to recover any appreciable amount of the energy liberated at the same time. Thus, in the Hanford production reactors, the only application being considered is for heating buildings. However, the energy used in this manner actually represents only a small proportion of the total released in fission of the uranium-235. Consequently the production reactors in operation at the present time are wasteful from the long-range standpoint of the utilization of nuclear energy.

12.36. As mentioned in Chapter I, reactors for the production of plutonium owe their existence to the use of this material in weapons. Assuming that this is sufficient justification for such reactors, some consideration may be given to the general problem of their design. The basic requirement of a production reactor is that neutron losses, by parasitic capture in moderator, coolant, structure, etc., and by escape from the system, be kept at a minimum, so that the conversion ratio (§ 1.151) be as large as possible. According to equation (1.151.1), the

maximum value of the conversion ratio, C, if there were no loss of neutrons, would be $\eta - 1$, where η is the number of fission neutrons produced for each neutron absorbed in the fissionable material. If the latter is uranium-235, the maximum value of C is thus 1.1, since η is 2.1 (Table 1.7). However, this argument does not take into account the gain in the number of neutrons due to the fast-fission effect (§ 3.131). The maximum possible conversion ratio is thus $\eta\epsilon - 1$, where ϵ is the fast-fission factor in the given system. The relationship between the conversion ratio and the nuclear and other characteristics of the reactor can be derived as follows. The result is applicable to all cases, not merely production reactors, in which conversion (or regeneration) occurs.

The Initial Conversion Ratio

12.37. Consider a fuel consisting of uranium-235 and uranium-238 only. For each thermal neutron causing fission in uranium-235, there will be $1 + \alpha$ neutrons captured by the uranium-235, where α is the ratio of nonfission captures to fissions in this nuclear species (§ 1.105). At the same time, $(1 + \alpha)\Sigma_{238}/\Sigma_{235}$ neutrons will be captured by uranium-238, with the ultimate formation of the same number of plutonium nuclei. Writing r for the isotopic ratio N_{235}/N_{238} (cf. § 1.108), this number may be expressed as $(1 + \alpha)\sigma_{238}/r\sigma_{235}$, where σ_{238} and σ_{235} are the absorption (nonfission + fission) cross sections of uranium-238 and uranium-235, respectively, for thermal neutrons.

12.38. As a result of the fission capture of a thermal neutron by uranium-235, referred to above, ν fast neutrons will be liberated, and they will produce $\nu\epsilon$ fast neutrons, where ϵ is the fast-fission factor. As these neutrons slow down to the uranium-238 resonance region, some will leak out from the reactor, and the nonleakage probability is given by $e^{-B^2\tau}$, where B^2 is the buckling of the reactor and τ is the Fermi age of the resonance neutrons (cf. § 3.120). The number of neutrons reaching the uranium-238 resonance region is thus $\nu\epsilon e^{-B^2\tau}$. If p is the resonance escape probability, the number of resonance neutrons captured by uranium-238 is then $\nu\epsilon(1 - p)e^{-B^2\tau}$, and this is the number of plutonium-239 nuclei that will ultimately be formed.

12.39. The foregoing results may thus be summarized as follows:

Number of plutonium-239 nuclei formed as a result of capture of thermal neutrons $= (1 + \alpha)\dfrac{\sigma_{238}}{r\sigma_{235}}$

Number of plutonium-239 nuclei formed as a result of capture of resonance neutrons $= \nu\epsilon(1 - p)e^{-B^2\tau}$

Number of uranium-235 nuclei destroyed $= 1 + \alpha$.

Since the conversion ratio is the ratio of the total number of fissionable (plutonium-239) nuclei formed to the number (of uranium-235) destroyed, it follows that

$$C = \frac{(1 + \alpha)\dfrac{\sigma_{238}}{r\sigma_{235}} + \nu\epsilon(1 - p)e^{-B^2\tau}}{1 + \alpha}$$

$$= \frac{\sigma_{238}}{r\sigma_{235}} + \eta_{235}\epsilon(1 - p)e^{-B^2\tau}, \qquad (12.39.1)$$

where η_{235}, equal to $\nu/(1 + \alpha)$, is the number of fission neutrons released per neutron absorbed in the uranium-235 (§ 1.105). It should be mentioned that the conversion ratio calculated here is for a fuel consisting of uranium-235 and uranium-238 only; it is consequently referred to as the "initial conversion ratio." As operation of the reactor proceeds, fissions occur in plutonium-239, as well as in uranium-235, and neutron capture in the resonance region is not restricted to uranium-238. The conversion ratio thus tends to decrease.

Example Suppose that in a natural uranium-graphite reactor p is 0.905 and ϵ is 1.03, calculate the expected initial conversion ratio of uranium-235 to plutonium-239.

In a reactor of this type, B^2 is about 10^{-4} cm^{-2} (see example following § 3.190); the age of thermal neutrons is about 400 cm^2, and the value for resonance neutrons may be estimated to be roughly 300 cm^2, so that $B^2\tau$ is approximately 3×10^{-2}. The nonleakage factor $e^{-B^2\tau}$ is thus close to 0.97. For uranium-235, η is 2.1 and the cross sections σ_{235} and σ_{238} are 650 barns and 2.80 barns, respectively; for natural uranium r is 0.00717, and so, from equation (12.39.1),

$$C = \frac{2.80}{(0.00717)(650)} + (2.1)(1.03)(1 - 0.905)(0.97)$$

$$= 0.60 + 0.20 = 0.80.$$

General Design Considerations

12.40. It may be seen from equation (12.39.1) that, for a given fuel, the first term on the right is constant; a large conversion ratio can thus be achieved by an increase in the fast-fission factor, a decrease in the resonance escape probability, and an increase in the nonleakage probability. In reactors based on natural uranium as the fuel and graphite as moderator there is little scope for changes in ϵ and p that will result in an increase in the conversion ratio. As seen in § 3.180, the maximum value of the infinite multiplication factor for natural uranium-graphite is only about 1.065, and so there is not very much that can be done in the way of design changes in order to increase the initial conversion ratio appreciably above 0.8.

12.41. With heavy water as moderator and natural-uranium metal as fuel, the infinite multiplication factor can be as large as 1.25. In this case an appreciable decrease in the resonance escape probability is permissible without making the infinite multiplication factor so close to unity that the reactor becomes impractically large. The conversion ratio in a natural uranium-heavy water system can thus be made larger than when graphite is the moderator, with the same (or smaller) fuel inventory.

12.42. Another possibility, irrespective of the nature of the moderator, is to increase the infinite multiplication factor by a slight enrichment of the uranium-235 content of the fuel. Although an increase in the isotopic ratio, r, will cause some decrease in the conversion ratio, due to the decrease in the first term on the right of equation (12.39.1), this can be more than offset by the decrease in the resonance escape probability while still maintaining the multiplication factor in excess of unity. With slightly enriched fuel it is possible to use ordinary water as moderator, and by suitable arrangement of the fuel elements very high values of the fast-fission factor, together with a low resonance escape probability, can be realized, thus leading to large conversion ratios. Whether the use of enriched fuel is worth while depends upon the value of the additional plutonium produced as compared with that of the uranium-235 required for enrichment, on the one hand, and upon the saving that may be realized by using ordinary water as moderator, on the other hand. Another factor which must be taken into consideration is the size of the reactor. With slightly enriched uranium as fuel and ordinary water as moderator, it is possible to build reactors of moderate size, thereby decreasing constructional costs.

Water-cooled Production Reactors

12.43. It is well known that in the Hanford production reactors the moderator is graphite and the coolant is ordinary water, with natural uranium as fuel.* The infinite multiplication factor in these reactors cannot be much greater than unity, at best, and the fuel-moderator arrangement is, of necessity, such as to give a fairly high value, e.g., 0.9, of the resonance escape probability. The attainment of a larger conversion ratio, approaching unity, is therefore not possible. Some improvement in the conversion ratio may be expected in the Savannah River reactors, which use heavy water as the moderator. An ideal design, for a heavy-water moderated production reactor, would involve cooling the fuel by circulating the moderator through a heat exchanger, as in the CP-3′, CP-5, and similar reactors, so that there is no ordinary water within the core. Although this would mean an appreciable increase (probably 15 to 25 per cent) in the total amount of heavy water required, there would be a gain in the conversion ratio.

12.44. If ordinary water is the internal coolant in a reactor in which essentially all the neutron moderation occurs either in heavy water or in graphite, the system may have an element of instability, which might possibly constitute a hazard. This is characteristic of any thermal reactor employing an internal (essentially nonmoderating) coolant that is a better neutron absorber than the moderator. If the flow of coolant should stop and its temperature rise, e.g., to the point where vapor is produced and the coolant is ejected, the rate of neutron capture

* H. D. Smyth, "Atomic Energy for Military Purpose," U. S. Government Printing Office, 1945, §§ 7.15 to 7.18.

will decrease, due partly to the decrease in density and partly to the decrease in capture cross section. Thus, additional neutrons may be available in the reactor core and the system may become supercritical. This can result in excessive local temperatures and damage to the reactor.

12.45. The chances of an accident can be largely obviated by associating a scram signal with the coolant water flow, so that the reactor is rapidly shut down when there is a notable decrease in the flow rate (cf. Table 6.4).* Nevertheless, it was the possibility of a mishap that led to the choice of air or carbon dioxide, rather than water, as coolant in the British plutonium production and power reactors (§ 8.230). The increased cost of pumping is repaid, to some extent, by the decreased neutron absorption.

12.46. It should be emphasized that the possibly hazardous situation considered above, when using ordinary water as reactor coolant, can arise only when the design is such that relatively little moderation of the neutrons occurs in the water. By the use of slightly enriched uranium as fuel it is possible to construct a reactor in which a fair proportion (or all) of the neutrons are slowed down in the water coolant while the others are moderated in graphite or heavy water. The reactivity of such a system would not be increased by a cessation in the flow of coolant.

POWER REACTORS

Mobile Reactors

12.47. Reactors for power production may be considered in two main categories, although there are cases of overlap as well as of different types within each group. The two classes are (1) mobile reactors for use in aircraft, submarines, surface ships, etc., and (2) stationary reactors for central-station, electrical power plants. Possessing some of the characteristics of both is a transportable reactor, capable of being readily assembled from parts which can be carried by air, and intended for stationary installation at a remote location, e.g., a military base.

12.48. The chief requirement of a mobile reactor, especially for propulsion of an aircraft or submarine, is that it should be relatively small and not excessively heavy. This means that the fuel must be appreciably enriched in fissionable material; its composition may be expected to approach pure uranium-235 or plutonium-239. The high cost of the fuel will not be very significant, in view of the advantages to be gained from the ability to operate for long periods without refueling.

12.49. With enriched fuel, there is considerable scope in choice of moderator and coolant. Water can act in both capacities simultaneously, and although

* In all reactors operating at appreciable powers, there must be provision for removal of the heat liberated by radioactive decay of the fission products after shut-down (§ 2.180). Otherwise, there is a possibility that the reactor may be damaged.

it has disadvantages, as will be seen shortly (cf. also § 8.234), it was chosen for the Submarine Thermal Reactor (STR), installed in the *U.S.S. Nautilus*. To attain the high temperatures necessary for satisfactory thermodynamic efficiency of the steam turbines, the water in the reactor must be under considerable pressure, so that a strong (and heavy) pressure vessel is required. In any event, assuming suitable materials to be available, the operating temperature is limited to something considerably below the critical value for water (705°F).

12.50. In order to obtain steam at high temperatures without having the reactor under pressure, the Submarine Intermediate Reactor (SIR), for the *U.S.S. Sea Wolf*, employs liquid sodium as coolant and beryllium as moderator. The relative amounts of beryllium and fissionable material are such that a large proportion of the fissions are caused by neutrons with energy in excess of the thermal value, i.e., in the "intermediate" energy range.

12.51. The Army Package Power Reactor* (APPR), for installation at a remote base where fuel costs are extremely high, is to be mobile in the sense that all the components can be carried in an aircraft. The heat-power level of the reactor is to be 10,000 kw and the electrical output 1700 kw. Refueling will take place at intervals of 12 months; it can be readily calculated that for continuous operation during this period the consumption of fissionable material will be about 3.6 kg, or roughly 8 lb.

12.52. The provisional specifications indicate that the fuel elements will consist of highly enriched uranium clad in stainless steel, and that water under a pressure of 1200 psi will be both moderator and coolant. A 2-in. thick "thermal shield," made of stainless steel, will be located between the reactor core and the walls of the pressure vessel, in order to reduce the thermal stress in the latter by absorbing gamma radiations from the core (see example following § 11.97).

12.53. The water will enter the pressure vessel at 432°F and will leave at 450°F. Circulation of this water through a heat exchanger (steam generator) will produce saturated steam at about 380°F and 200 psia. The over-all efficiency for electrical power production is expected to be about 17 per cent, but the net efficiency will be less because of pumping requirements.

Central-station Power Reactors

12.54. In the design of a reactor for central-station power, economic considerations, which are not as significant as size for mobile reactors for military purposes, become of prime importance. Although there are now no strict limitations on size or weight, excessive size would undoubtedly prove to be uneconomical because of the cost of the shield, reactor building, and pressure vessel (if used), to say nothing of the large fuel inventory that would be required. Many stationary power plants of the near future will probably use slightly enriched uranium, e.g., containing 1.2 to 2.0 per cent uranium-235, as fuel. In this way, reactors of reasonable size will be possible. Which moderator or coolant,

* *Nucleonics*, **12**, No. 3, 76 (1954); **12**, No. 10, 22 (1954); **13**, No. 5, 24 (1955).

fuel-moderator system, or method of heat removal will prove best is still an open question, and it is the purpose of the reactor experiments now under way to help supply a possible answer. The arguments for and against some of the design variables will be discussed below.

12.55. One aspect of the experimental program is the construction of the full-scale (60,000-kw) electrical power plant at Shippingport, Pa. (see § 1.37), using a reactor of 264,000 kw to supply the necessary heat. The reactor, called the Pressurized Water Reactor (PWR), will use slightly enriched fuel and water at about 2000 psi as moderator and coolant (for further details, see § 13.80). Although it is generally believed that this plant as designed will not produce electrical power that is competitive with the lowest cost coal-fired plants, the particular system was chosen largely because of the technological experience with water-cooled and -moderated reactors, e.g., the STR and MTR. It is expected that operation of the PWR and its associated equipment will provide information which will contribute to the ultimate design of economic nuclear power plants.

Dual-purpose Plants

12.56. There has been some interest in recent years in dual-purpose plants, mentioned in Chapter I, designed to produce both nuclear power and plutonium for weapons.* Actually, any reactor which contains uranium-238 will be a converter, in the sense that some plutonium-239 will be produced. Normally, however, the plutonium would remain in the reactor and would contribute to the fissions. Eventually, if conditions, e.g., the total amount of fissionable material present, accumulation of fission products, and mechanical stability of the fuel elements, permitted, the amount of plutonium-239 present would attain a limiting (or equilibrium) value, as noted in § 2.205. The plutonium would then be consumed, mainly by neutron capture and fission (and to a very minor extent by radioactive decay), as fast as it was formed from uranium-238. In the processing of the spent fuel elements the residual plutonium would, of course, be recovered, but it would probably be too impure for use in weapons, due to the presence of higher isotopes of plutonium.

12.57. In a dual-purpose reactor it would be necessary to remove the fuel elements and extract the plutonium long before their usefulness in the reactor, from the standpoint of energy production, had ceased. The limit thereby set upon the extent of reactor operation before refueling might affect the design. In the dual-purpose reactor, the emphasis would be on ease of insertion and removal of fuel elements, as well as on simplicity of processing. Further, the reactor design chosen to give a high conversion ratio would not necessarily be the same as that for maximum efficiency for power production. In a power-only

* For discussions of the relative merits of single- and dual-purpose nuclear power plants, see C. Starr, *Nucleonics*, **11**, No. 1, 62 (1953); C. A. Hochwalt and P. N. Powers, *ibid.*, **11**, No. 2, 10 (1953); G. L. Weil, *ibid.*, **11**, No. 4, 12 (1953); K. Cohen, *ibid.*, **11**, No. 5, 10 (1953).

system, on the other hand, long life of the fuel elements would be a prime consideration.

12.58. There is little doubt that electric power from a dual-purpose nuclear plant would be more expensive than from a power-only reactor, and the former could be justified only if weapons-grade plutonium could be sold at a premium price.* It appears that the production reactors at present in operation or under construction, at Hanford and Savannah River, can supply all the present requirements of the United States for this material.†

12.59. It may be noted that, although a power-breeder is, in a sense, a dual-purpose reactor, it differs in some respects from a reactor which is designed to produce plutonium-239 from natural-uranium fuel. It will be seen in due course that in most cases power-breeder reactors will be designed as two-region reactors; that is to say, the core, in which most of the energy is produced, will be separate from the breeding region. Thus, processing of the blanket can be made independent of the operation of the core.

Maximum Operating Power

12.60. In the subsequent sections it will be assumed that the reactor under consideration is intended for power production and the design variables of such a system will be discussed. The first decision to be made is concerned with the maximum operating (or heat) power level. For mobile or transportable reactors, this may range from 10,000 kilowatts, i.e., 10 megawatts (10 Mw), as for the APPR, to about 300 Mw, yielding some 100,000 shaft horsepower, for the propulsion of a large ship. For a central-station power plant, the heat output of the reactor might range from 100 to 2000 Mw, corresponding to an electrical capability, at full load, of perhaps 20,000 to 500,000 kw. This is, roughly, the operating range of conventional electric-power plants at the present time, although a few have outputs above 1000 Mw.

12.61. The power level decided upon is the maximum rate of heat production for which the reactor is designed to operate continuously for a long period, without the risk of damage to the components or danger to personnel. Operation below the maximum power is, of course, always possible by suitable adjustment of the particular heat-removal system embodied in the reactor design. It should be mentioned that, although the maximum power level is more or less fixed in advance, it may be increased somewhat as the operation proceeds. For example, a change in the permissible temperature, due to improvements in the fuel elements, may perhaps make possible a more efficient or less expensive heat exchanger (boiler) for steam production or turbogenerator system for electricity.

* It is reported that the first British (Calder Hall) power reactor is to be of the dual-purpose type. It will be a natural uranium-graphite reactor cooled by carbon dioxide under pressure (see *Atomics*, **5**, 174, 275 (1954)). It should be pointed out, however, that the average cost of electric power in the United Kingdom is appreciably greater than in the United States, and the plutonium-production facilities are less extensive.

† From speech by T. E. Murray, summarized in *Nucleonics*, **12**, No. 10, 77 (1954).

THE NUCLEAR SYSTEM

Neutron Energy Spectrum in Reactors

12.62. The purpose of the reactor and its maximum power output, treated above, are, in some respects, related to the nuclear system. The present section, however, will deal more specifically with nuclear matters, such as neutron energy, components of the reactor (fuel, moderator, and reflector materials), methods of control, stability, and criticality.

12.63. The design of the nuclear system and the neutron-energy spectrum in the reactor, i.e., the neutron-energy region in which most of the fissions occur, are interdependent. In general, the energy regions are described broadly as fast, intermediate, and thermal, and one of these must be selected as a first step. Each energy range has its advantages and disadvantages, as will be seen below.

Fast Reactors

12.64. The outstanding argument for fast reactors, in which most of the fissions are produced by neutrons of energy in excess of about 0.1 Mev, is that parasitic capture and nonfission absorption of neutrons by the fuel are much less significant than at thermal energies. High conversion ratios (or breeding gains) are thus possible, as mentioned in Chapter I. Without such breeding (or efficient conversion), the future of the nuclear-power industry would appear to be somewhat limited. It is true that breeding of uranium-233, from thorium-232, is expected to prove possible with neutrons in the thermal-energy range, but the world supply of economically recoverable thorium is believed to be much less extensive than that of uranium (§ 1.5).

12.65. The small values of the absorption cross sections of most elements for fast neutrons and the high concentration of fissionable material in the core make possible a wider choice of constructional and cladding materials in a fast reactor. The designer is no longer restricted to using aluminum or zirconium to minimize neutron losses and to achieve a large conversion ratio. Stainless steel and other alloys possessing good mechanical properties and resistance to corrosion and erosion can now be employed without seriously affecting the neutron economy. However, the coolant must be restricted to one that does not cause appreciable slowing down of the fast neutrons. Exposure to neutrons of high energy may cause severe radiation damage to certain constructional materials, thus precluding their use.

12.66. Fission-product poisoning in a fast reactor is also less significant than in a thermal reactor, because of the smaller cross sections; consequently, if the effects of radiation permit, a greater degree of burn-up of the fuel elements may perhaps be possible. In any event, the practical limitation to the neutron flux, due to the xenon poisoning effect (see Chapter IV), does not apply to a fast reactor.

12.67. Among the drawbacks of fast reactors appear to be their high fuel inventory and small size. In order to avoid appreciable slowing down of the neutrons, in both inelastic and elastic collisions, it is necessary that the fuel in a fast reactor contain a large proportion of fissionable material. Further, because of the much smaller fission cross section, the critical mass is larger for fast than for slow reactors. There is thus an unfortunate combination of circumstances in which there will be a demand for fissionable material, for the cores of breeder reactors, in the earlier stages of the nuclear industry, when such material is in short supply.

12.68. Due to the absence of a moderator and of other materials of low mass number, the core of a fast reactor is usually small; thus, in the Los Alamos fast reactor, the plutonium core, including coolant channels, was a cylinder about 6 in. in diameter and 6 in. high, and the core of the EBR has been described as being "as big as a football" (§ 1.9). The small size means a small heat-transfer area, so that removal of large amounts of heat by a coolant is difficult. The situation can be greatly improved by using fuel elements consisting of 80 per cent or so of fertile material, e.g., uranium-238 or thorium-232, and the remainder of a fissionable species. Nevertheless, the actual power (or specific power) of a fast reactor is limited by the possible heat removal from a structure of fairly small dimensions. Because of the moderating effect of water, either ordinary or heavy, it is necessary that a liquid metal or other nonmoderating coolant be used for a fast reactor.

12.69. It is believed that the maximum specific power so far attained in a fast reactor is about 50 kw/kg of fissionable material, although there are prospects for larger values, e.g., 500 to 1500 kw/kg in EBR-2. A high specific power is desirable for two reasons, at least; first, to minimize the fuel investment charge per kw-hr of electricity and, second, to decrease the doubling time in a breeder reactor (§ 1.157).

Intermediate Reactors

12.70. If an attempt is made to increase the rate of heat removal from a fast reactor by introducing more coolant, the size of the core will be increased and some slowing down of the neutrons will be inevitable. This could be compensated, to some extent, by adding more fissionable material, which might be necessary, in any event, to maintain criticality, but then there would be no gain in the specific power. As an alternative, the reactor might be operated in the intermediate neutron-energy region. Whether this is worthwhile or not will depend on the variation of capture and fission cross sections and, hence, of the breeding gain with neutron energy. If many of the advantages of a fast reactor are lost, there would appear to be no special merit to an intermediate reactor.

12.71. Only one intermediate reactor, the SIR referred to in § 12.50, is in the active planning or construction stage. Since some beryllium is used as moderator, although not sufficient to thermalize most of the neutrons, it must be presumed that fissions are caused mainly by neutrons in the low-intermediate range, i.e., nearer to thermal than to fission energies. The critical mass of fissionable material in such a reactor would probably be less than in a fast reactor, because of the increase in the fission cross section, so that it could be designed to have a higher specific power. Values of 2000 kw/kg or more, approaching those considered possible for slightly enriched thermal reactors, are believed to be attainable with intermediate reactors. However, the intermediate reactor would have the advantage in respect to fission-product poisoning. If the neutron-energy range were moderately high, it could be above that at which xenon-135 is a significant poison. The problems of neutron-flux limitation and xenon override would thereby be eliminated.

12.72. Until some experience is gained in the operation of an intermediate reactor, it is not possible to state categorically whether or not it is superior to a thermal reactor for power production. Although breeding may prove feasible in the intermediate neutron-energy range, it is doubtful whether the breeding gain would be as large as for a fast reactor. The future of the intermediate reactor is thus in doubt and the results of the operation of the SIR will be awaited with interest.

Thermal Reactors

12.73. Apart from the requirements of neutron economy, there are fewer restrictions upon the design of a thermal power-reactor than on the types discussed above. Any form of fuel, from natural uranium to relatively pure fissionable material, can be used, in principle, and the ratio of fissionable nuclei to those of the moderator can often be varied considerably (cf. Fig. 3.13). This permits a wide range of sizes, and very small or very large thermal reactors are possible. The increased size facilitates total heat removal, and specific powers as large as 4000 kw/kg or more are attainable, when the fuel is highly enriched material.* When slightly enriched or natural uranium is the fuel, the specific power may be 2000 kw or less per kg of fissionable material; but the cost of a given mass of uranium-235 in the unseparated (in slightly enriched or natural uranium) form is much less than when separated. In addition the conversion of uranium-238 to fissionable plutonium-239 which occurs in a natural uranium (or moderately enriched) thermal reactor contributes to the over-all economy.

12.74. Some disadvantages, e.g., increased cost of shielding, buildings, etc., are associated with the larger size of thermal reactors, although the increase in size is not significant for reactors employing highly enriched fuel. In any event, the larger size greatly simplifies many of the auxiliary aspects of reactor design, such as coolant entry and exit ducts, mechanism for moving control rods, instrumentation, etc.

* The MTR, with a power of 30,000 kw has about 4 kg of uranium-235 in its core; its specific power is thus close to 7500 kw/kg of uranium-235.

12.75. Provided the reactivity is well below the prompt-critical value (§ 4.46), there should be no essential difference between the ease of control of thermal, intermediate, or fast reactors. The effective neutron lifetime in the reactor is then determined by the delayed neutrons. But if, due to accidental circumstances, the reactor should "run away," and the reactivity should increase beyond prompt critical, the chances of effective automatic corrective action being taken are greater with a thermal reactor than with the other types. This is because the prompt-neutron lifetime in a thermal reactor is 10^{-4} to 10^{-3} sec, but in a fast reactor it is only 10^{-7} sec or less. It should be recalled (Table 2.10), too, that the fraction of delayed neutrons is less for plutonium-239 than for uranium-235; hence, in a fast reactor using plutonium as fuel, e.g., a fast breeder-reactor, the prompt-critical reactivity will be attained more easily than in one in which uranium-235 is the fuel.

12.76. Perhaps the outstanding drawback to high-flux, thermal reactors is the limitation set by xenon poisoning and the ability to start up again after a short shut-down. It is true this problem can be partially overcome by building excess reactivity, in the form of additional fuel, into the reactor, or by refueling after a shut-down, but these procedures may be uneconomic (see § 6.34). Another solution, to be examined more fully later, is to employ a liquid fuel system, so that portions can be "bled" off continuously for removal of fission products.*

12.77. Although the situation may change as more experience is obtained in reactor operation, it appears that, at the present time, most designers favor thermal reactors for both mobile and central-station power production. Only when breeding of plutonium-239 is an important desideratum might fast reactors be preferred.

REACTOR COMPONENTS

Natural-Uranium Reactors

12.78. The reactor components to be reviewed here are primarily the fuel, moderator, and reflector. Some preliminary consideration will also be given to the nature of the coolant, since this is related to the choice of the other components.

12.79. The fuel for a thermal reactor can be natural uranium or uranium of any degree of enrichment in uranium-235. Even depleted uranium, i.e., with less than the normal proportion of uranium-235, could be utilized, to a limited extent, in special cases. Plutonium-239, either in the relatively pure state or in combination with a fertile material, could be used as fuel. No thermal reactor design based on plutonium as fuel has yet been considered seriously in the United States, but ultimately reactors of this type should receive attention. Similarly,

* It is probable that many power reactors, not using highly enriched fuel, will operate at thermal fluxes appreciably less than 10^{14} neutrons/(cm^2)(sec). The xenon poisoning problem will then not be serious.

the fissionable material could be uranium-233 when sufficient quantities of this isotope are available.

12.80. The choice of moderator is partly dependent upon the composition of the fuel. If the latter is natural uranium, ordinary water cannot be used as moderator. Even if a critical system of natural uranium and water could be attained, which is by no means certain, it would be too large to be of any practical value.* From the nuclear standpoint, the best moderator for use with natural uranium fuel is heavy water, since it requires the smallest fuel inventory and permits a critical reactor of moderate size. For example, a system with about 2.5 tons of natural uranium, in the form of long rods, and 6.5 tons (1500 gal) of commercial heavy water can become critical and support a fission chain reaction. This could be attained in a cylindrical tank about 6 ft in diameter and 9 ft high. Even allowing for additional fuel and moderator, required at high temperature for criticality and for heat removal, the dimensions would not be unreasonable for a pressurized system, such as is necessary to realize temperatures that would give an acceptable thermodynamic efficiency. In order to economize in the total amount of the expensive heavy water required, graphite could be used as reflector, with some increase in the core size. Beryllium and beryllium oxide are excellent reflectors, but the installed cost would probably be as large as that of the heavy water saved.

12.81. Another argument in favor of heavy water as moderator, with natural uranium as fuel, is that a higher conversion ratio is possible than with other moderators (see § 12.43). As a result, the "burn-up," defined as the proportion (or percentage) of the total fuel† that is consumed before the reactor must be shut down for recharging, can be larger when heavy water is the moderator, provided, of course, that radiation damage to the fuel elements is not a limiting factor. The decrease in frequency of replacing and processing the fuel elements would be an important contribution to economical power production. Further, the relatively long thermal-neutron lifetime in heavy water (see Table 3.6), due to its small capture cross section, is advantageous. The reactor period for fairly large reactivities is longer than for other moderators and so heavy-water reactors are safer (cf. § 4.38).

12.82. There is no doubt that if heavy water were available at a reasonable cost, e.g., about \$20 per lb instead of the quoted commercial price of \$80 per lb, natural uranium-heavy water reactors would be of greater interest for power production.‡ The investment in heavy water is increased some 15 to 25 per cent,

* Cf. R. Persson, *Nucleonics*, **12**, No. 10, 26 (1954).

† In defining "burn-up," the term "fuel" is used in the general sense referred to in § 1.91, i.e., including any fertile material that may be present.

‡ The design and economics of a power reactor of this type have been considered by the Nuclear Power Group, consisting of American Gas and Electric Service Corp., Bechtel Corp., Commonwealth Edison Co., Pacific Gas and Electric Co., and Union Electric Co. of Missouri; see *Nucleonics*, **12**, No. 7, 50 (1954); **12**, No. 10, 67 (1954); also T. G. LeClair, Atomic Industrial Forum Report, July 1954, p. 58.

because of the volume held in the external heat exchanger, if, as is highly desirable, this liquid also serves as the coolant. It has been suggested that some reduction in cost could be achieved by using ordinary water as coolant, but this may lead to the introduction of an undesirable element of instability into the reactor (see § 12.44), as well as to an increased loss of neutrons. A more practical solution might prove to be a boiling heavy-water reactor; the amount of the moderator-coolant outside the reactor would then be a smaller proportion of the total.

12.83. Other materials that can be used as moderators in natural-uranium reactors are beryllium, beryllium oxide or carbide, and graphite. Because of the quantity of material required (50 to 100 tons for the core and somewhat more for the reflector), it is uncertain whether either beryllium metal or any of its compounds, which are relatively costly to produce and fabricate (see Chapter VIII), will find extensive application in natural-uranium systems in the near future. This leaves graphite as the only remaining immediate practical possibility.* However, as seen earlier, a natural uranium-graphite reactor is inevitably large — a minimum volume, excluding the reflector, being about 8000 ft³ — and this introduces some difficulties in the design of a reactor for power production.

12.84. For high-temperature operation, such as is essential for a power reactor, the probable coolants for a natural uranium-graphite system are (1) a gas at high pressure, (2) pressurized water (ordinary or heavy), or (3) a liquid metal.† Some of the disadvantages of a gaseous coolant have been mentioned in Chapter VIII, and reference will be made here to the special problems associated with power reactors. The use of a gas as coolant would require either a pressure vessel large enough to contain the reactor core or pressurization only of the cooling channels surrounding the fuel elements. Each of these alternatives would involve difficult, but not insuperable, constructional problems. The design for a helium-cooled, graphite-moderated, natural uranium, dual-purpose reactor was included in the initial proposals made by one of the industrial groups referred to in § 1.23, but it was subsequently discarded (cf. § 8.232). As far as nuclear-reactor designers in the United States are concerned, the general consensus appears to be that gas cooling is not one of the most promising paths to the attainment of economic nuclear power. The British (Calder Hall) power reactors, with natural uranium as fuel, are to be cooled by carbon dioxide gas, but this choice was determined partly, at least, by safety considerations and partly because of experience with the air-cooled (Windscale) production reactors.

12.85. If either ordinary or heavy water were the coolant, it would also be necessary to have a pressurized system and the mechanical difficulties would be

* Some designers are of the opinion that before long beryllium or beryllium oxide will be able to compete economically with graphite as moderator in power reactors.

† Another possibility would be an organic compound or mixture (see § 8.255).

similar to those with a gaseous coolant. With ordinary water as coolant, and another material as moderator, the loss of neutrons by capture in the water would require that the reactor core be even larger than indicated above.

12.86. The difficulties associated with a pressurized coolant could be avoided by using a liquid metal. Sodium or NaK may not be suitable for natural-uranium reactors because of their relatively large capture cross sections for thermal neutrons, but either the lead-bismuth or the lead-magnesium eutectic alloys (see Table 8.18) might be possible. However, relatively little is yet known about the technology of these materials at high temperatures. It is of interest to recall that cooling by molten bismuth was given serious consideration when the Hanford production reactors were being designed. Although this coolant appears to be very promising from the standpoint of utilizing the heat liberated in the reactors, it was realized that solution of the technological problems involved would require considerable research and development.*

Slightly Enriched Uranium Reactors

12.87. By employing even slightly enriched uranium as fuel, the flexibility of the reactor system is very greatly increased. This is due to the significant increase in the multiplication factor resulting from a moderate increase in the uranium-235 content of the fuel. For example, if the uranium contains 1.2 per cent, instead of the normal 0.712 per cent, of the lighter (fissionable) isotope, it can be readily shown from equation (1.107.1) that η is increased from 1.3 to nearly 1.5. Further, enrichment of the fuel may be expected to cause some increase in the thermal utilization, for a given moderator, since a larger fraction of the thermal neutrons are captured in the fuel. Even if the resonance escape probability, the thermal utilization, and the fast-fission factor, remain essentially unchanged, an increase in the uranium-235 enrichment to 1.2 per cent will result in a 15 per cent increase, at least, in the infinite multiplication factor.

12.88. The increase in the multiplication factor accompanying this relatively small enrichment of the fuel makes possible the design of a reactor with water as moderator and coolant. In a heterogeneous natural uranium-water system, an infinite multiplication factor of about 0.97 has been observed, so that with uranium enriched to 1.2 per cent as fuel, the value of this factor may be expected to be close to 1.15. Because of the small migration area of neutrons in ordinary water (see Table 3.5), it is possible with such a relatively high multiplication factor, to design a reactor of reasonable size, comparable with that for a natural uranium-heavy water system. The PWR, for example, which is to use slightly enriched uranium metal as fuel, and pressurized water as moderator and coolant,

* H. D. Smyth, *op. cit.*, § 6.43. It now appears that, although the lead-bismuth eutectic has a higher cross section for neutron capture, it has better heat-transfer properties than bismuth alone.

is expected to contain somewhat more than 15 tons of fuel in the core, in a cylindrical arrangement about 6 ft in diameter and 7.5 ft high.*

12.89. Whether an enriched uranium-water reactor for power production is economically sound or not depends to some extent (but not entirely) on the cost of uranium-235 relative to the savings resulting from the use of a cheap moderator and coolant. In this connection a reasonably good comparison is an enriched uranium-water reactor with one employing natural uranium as fuel and heavy water as moderator. To enrich uranium from the normal 0.712 per cent of uranium-235 to 1.2 per cent, for example, would require about 10 lb of this material per ton of uranium fuel. For a reactor containing 10 tons of fuel, the cost of the uranium-235, at $20 per gram,† would be about $900,000 per loading. The price of heavy water for a similar reactor, at $80 per lb, would be at least $3,000,000 so that, on the basis of about 10 per cent annual capital charges and 5 per cent annual loss of heavy water, the costs would be comparable if the enriched reactor had a fuel-cycle time of just over 2 years, i.e., the fuel would have to be completely replaced once every 2 years or so.

12.90. Actually, the cost factors considered above are only part of the problem. For example, the value of the heavy water means that special precautions would have to be taken against leaks in pumps, pipes, etc., and this would require a more expensive plant. A clear-cut choice between ordinary- and heavy-water reactors for power purposes cannot yet be made.

12.91. Another design, which requires the enrichment of the fuel, involves a heterogeneous uranium-graphite reactor with liquid sodium as the coolant. A reactor of this type would be of moderate size, it would not need to be pressurized, and it could operate at appreciably higher temperatures, and give a better overall thermal efficiency, than one using liquid water as coolant. The infinite multiplication factor of a system of 1.2 per cent enriched uranium with graphite as moderator, disregarding the coolant, could approach 1.25. Hence, there would be scope for cooling by liquid sodium, with its fairly large absorption cross section for neutrons, as well as for the use of stainless steel (or other resistant alloy) for cladding and structural purposes. A reactor of this type is the basis of the Sodium Reactor Experiment (SRE) included in the A.E.C.'s Five-Year

* C. H. Weaver, Atomic Industrial Forum Report, July 1954, p. 16; see also, *Nucleonics*, **12**, No. 7, 48 (1954). In the "Report on the Five-Year Power Reactor Development Program Proposed by the Atomic Energy Commission" (U. S. Government Printing Office, March 1954) it is stated that the fuel charge will consist of 15 to 20 tons of uranium containing 1.5 to 2 per cent of uranium-235.

† This figure for the cost of uranium-235 is frequently quoted in the open literature; see, for example, L. F. Hafstad, in the Joint Committee on Atomic Energy Report on "Atomic Power and Private Enterprise," U. S. Government Printing Office, 1952, p. 132; W. H. Zinn, *Nucleonics*, **10**, No. 9, 8 (1952). If 13,000 Btu/lb coal costs $7 per ton, the fuel value of uranium-235, assuming complete fission, i.e., with an energy release of 3.6×10^{10} Btu/lb, would be about $21 per gram.

Development Program.* It may be regarded as the first step toward the larger Sodium Graphite Reactor (SGR).†

12.92. For a given reactor type, it may be possible to increase the conversion ratio by adjusting the uranium-235 enrichment and the proportion of fuel to moderator. It can be seen from equation (12.39.1) that this can be achieved by increasing ϵ and decreasing p, but to maintain criticality it may be necessary to increase the concentration of uranium-235. However, such a change would also mean an increase in r, which would have the opposite effect on the conversion ratio. By proper balancing of the enrichment and the ratio of fuel to moderator, the conversion ratio may be increased, within certain limits, and also perhaps the burn-up, assuming adequate radiation stability of the fuel elements. It should be noted that the possible burn-up depends upon a variety of other factors, including the number of neutrons produced by the fission of higher isotopes of the elements present in the fuel and the loss of neutrons due to capture by fission (and other) products formed during reactor operation.‡

Highly (or Moderately) Enriched Uranium Reactors

12.93. With the exception of breeders or efficient converters, the use of highly enriched fuel, either uranium-235, uranium-233, or plutonium-239, in reactors is wasteful from the long-range, nuclear energy point of view. Nevertheless, such material is employed in special cases, as mentioned earlier, where mobility or compactness, is a prime consideration. With highly enriched fuel, there is a relatively wide choice of neutron energy, moderator (if required), coolant, and structural materials, so that the scope of the reactor designer is less restricted. The best combination of fuel, moderator, and coolant for power production has not been established; the promising possibilities, represented typically by the STR and SIR, respectively, are high-pressure water as moderator-coolant, and beryllium (or graphite) as moderator with liquid sodium as coolant. It must be admitted, however, that other systems, not yet tested, may ultimately prove to be preferable.

Fluid Fuel Reactors

12.94. Another approach to power-reactor design is the use of a fluid (instead of a solid) fuel. In this connection homogeneous aqueous solutions have been most extensively studied. The advantages (and disadvantages) of such systems will be examined shortly, and for the present only general aspects of

* C. Starr, Atomic Industrial Forum Report, July 1954, p. 11; see also, ref. to § 12.9, p. 12.

† See W. E. Parkins, *Chem. Eng. Prog. Symposium Series*, No. 13, **50**, 53 (1954). A power reactor of the same type was also proposed by the Monsanto Chemical Co. and Union Electric Co. (see ref. to § 1.23); a study of favorable design values is given by E. M. Mabuce and K. A. Hub, Monsanto-Union Electric Atomic Electric Project, Report AEP No. 99.

‡ For detailed treatments, see K. Bernstein, *Proc. Conf. on Nucl. Eng.*, Berkeley, 1953, A-35; T. M. Snyder, *ibid.*, C-11.

the core composition will be reviewed. A solution of a uranium salt of normal enrichment cannot be made critical in ordinary water, but a chain reaction can be maintained in heavy water. The maximum infinite multiplication factor that can be realized, e.g., with a solution of (natural) uranyl fluoride in heavy water, is about 1.18. By suitable enrichment of the uranium salt a critical homogeneous system can also be attained in ordinary water. Such solutions are used in the various forms of the Water Boiler reactor and in the Homogeneous Reactor Experiment (HRE). For a homogeneous breeder reactor, the loss of neutrons by absorption in ordinary water could not be tolerated and heavy water would have to be used as moderator.

12.95. In the HRE and in all recent forms of the Water Boiler, the fuel is initially highly enriched (90 per cent or more) in uranium-235. One reason is that this permits the use of a more dilute solution, i.e., one with a lower total concentration of uranium salts, which is less corrosive to certain metals. It should be recalled, however, that in the original (LOPO) form of the Los Alamos Water Boiler reactor, the fuel (uranyl nitrate solution) contained only 14.6 per cent of uranium-235 (§ 13.86).

12.96. The foregoing discussion of fluid fuel systems has referred, in particular, to cases in which the fuel consists of a uranium salt dissolved in water. However, other types of homogeneous reactors have been considered. Most of these are either in the conceptual design or speculative stages and they will be described below (§ 12.139, *et seq.*).

Power-Breeder Reactors

12.97. Reactors which produce useful power and, at the same time, breed fissionable material are expected ultimately to play an important role in the world's nuclear-power program. If plutonium-239 is to be bred from uranium-238, it may be necessary to employ fast reactors. As already seen, the core must then contain a high proportion of fissionable material, i.e., plutonium-239 in the present case. The conversion (or regeneration) process that results in breeding then takes place mainly in a blanket of natural (or depleted) uranium.

12.98. The presence of elements of low mass number in the core must be avoided as far as possible. For this and other reasons (§ 12.126), liquid metals are the most suitable coolants. In the EBR-1 the coolant is a sodium-potassium alloy (NaK) of low melting point, but it is proposed to use sodium alone in the EBR-2. Although the latter tends to slow down the neutrons more than NaK, its superior heat-transfer properties indicate that it may be preferable because of the problem of heat removal in a fast reactor. Heavy-metal coolants, such as lead-bismuth or lead-magnesium alloys, would have the advantage of not slowing down the neutrons, but they are less effective than sodium for heat-transfer purposes.

12.99. It may be noted that EBR-1 is not a true breeder, since it uses uranium-235 as fuel (see, however, § 13.121) while producing plutonium-239 in the "breeding" blanket. Its main purpose was to test the possibility of obtaining a conversion ratio of unity or more, and since a value of "at least unity" was attained with uranium-235, breeding should certainly be possible with plutonium-239 as fuel, because the latter has a larger value of η for fast neutrons. The EBR-2 will be a true breeder, for it is being designed so that in due course, if not at the outset, plutonium-239 will be used as the fissionable material.

12.100. Breeding of uranium-233 from thorium-232 is possible, in principle, in both thermal and fast reactors. Although the breeding gain would be expected to be larger in a fast reactor, the immediate interest is centered upon breeding in thermal reactors because of their other advantages (§ 12.73). Since there is no uranium-233 in nature, some of this fissionable isotope will have to be obtained by exposing thorium-232 to neutrons in existing reactors. Then, two different types of breeder design will be possible.

12.101. The first is a "one-region" (reflected) reactor, with a moderator, similar to the uranium reactors using natural or slightly enriched material, in which the fuel is a suitable mixture of uranium-233 and thorium. In this reactor, in addition to the release of energy which will be utilized for power purposes, there will be a conversion of thorium-232 into uranium-233. When the fuel elements are removed, either because of the accumulation of fission products and fissionable material or because of radiation damage, and processed, the resulting material will contain more uranium-233 than was initially present. Since it is not necessary, for a one-region breeder reactor, to separate the uranium-233 from the thorium, processing may be relatively simple, if methods can be developed for the removal of the fission products only.

12.102. The alternative is a "two-region" thermal (or fast) breeder, similar in principle to the fast breeder described above. In such a reactor, the core region would contain the fuel, having a high percentage of uranium-233 and a moderator; this would be surrounded by a breeding blanket of thorium-232, and a moderator. Separate chemical processing of the core and of the blanket would be necessary, and in the latter case the uranium-233 produced and the thorium-232 would have to be separated from one another and from fission products which will undoubtedly be present. However, the increased complexity of processing with the two-region, as compared with a one-region, thermal breeder will be compensated by the decrease in the frequency of processing the blanket. The latter need be processed only when the inventory of uranium-233 becomes uneconomically large.

12.103. An advantage of the two-region type of breeder is that it can be designed so as to have good neutron economy and moderate size. Since the fuel is highly enriched in uranium-233, the core can be made quite small, provided only that the heat removal requirements can be met. Apart from the advantage of moderate size, the total inventory of uranium-233 will probably be less than for a one-region breeder. The Homogeneous Thorium Reactor (HTR), which is part of the A.E.C.'s Five Year Program, is to be of the two-region type. In

order to facilitate heat removal and chemical processing, the fuel will be dissolved in a circulating solution. The solvent, acting also as moderator and, in a sense, as coolant, will be heavy water since there would be no breeding if ordinary water were used.

Summary

12.104. In the accompanying Table 12.2 there are summarized the main components of power reactors which are expected to show development in the not too distant future. (The symbol S.S. represents stainless steel or other resistant alloys.) Of course, many other reasonable combinations of fuel, moderator, and coolant are feasible, but they do not offer the same prospects for immediate success as those tabulated. As experience is gained, however, it is possible that the situation may change.

TABLE 12.2. COMPONENTS OF POSSIBLE POWER REACTORS

Fuel	Moderator	Reflector	Coolant	Structure
Natural uranium	D_2O	D_2O, Be, BeO, C	D_2O	Al, Zr
Slightly enriched uranium	H_2O, D_2O	H_2O, D_2O, etc.	H_2O, D_2O	Al, Zr, S.S.
	Be, BeO, C	Be, BeO, C	Na, H_2O	Zr, S.S., etc.
Highly enriched uranium	H_2O, D_2O*	H_2O, D_2O, etc.	H_2O	Al, Zr, S.S.
	Be, BeO, C	Be, BeO, C	Na	Zr, S.S., etc.
Uranium-235 or Plutonium-239†	None	U-238 blanket, + Be, BeO, C	NaK, Na, other liquid metal	S.S., etc.
Uranium-233‡	H_2O, D_2O	Th-232 blanket, + D_2O	H_2O, D_2O	Al, Zr, S.S.

* Homogeneous or heterogeneous.
† Fast breeder.
‡ Homogeneous thermal breeder.

Criticality and Related Problems

12.105. When the nature of the fuel and moderator have been decided upon, and a preliminary choice, at least, of coolant has been made,* it is possible to carry out preliminary criticality calculations. As seen in Chapter III, the procedure depends upon whether the fuel-moderator system is homogeneous or heterogeneous. With natural or slightly enriched uranium as fuel, the reactor will probably (though not necessarily) be heterogeneous, but with moderately or highly enriched fuel, a homogeneous system is possible. In small reactors containing highly enriched fuel, especially with ordinary water as moderator, the fuel elements generally have a high ratio of area to volume and, in addition, are close together. As far as the criticality calculations are concerned, such reactors, although strictly heterogeneous, can be treated as homogeneous. Consequently they are sometimes referred to as quasihomogeneous.

* A more detailed analysis of the coolant problem will be given below in the section dealing with the design of the heat-removal system.

12.106. For homogeneous thermal reactors there is a particular ratio of fuel to moderator which gives a critical system with a minimum mass of fissionable material (cf. Fig. 3.14). Similarly, there is a particular size and arrangement of fuel elements which give a maximum multiplication factor in a heterogeneous reactor. In a power reactor, it is very unlikely that these optimum design conditions, from the nuclear standpoint, will permit removal of heat at the rate required by the stipulated power of the reactor. It is necessary, therefore, to calculate the critical dimensions (and masses) of the fuel-moderator system for a variety of conditions; among the variables to be considered are the nature and quantity of coolant, if it is not the same as the moderator, and its effect on the thermal utilization.

12.107. At the same time as the criticality studies are being made, the thermal problems of the reactor will be investigated. Coolant temperatures and coolant velocity, and related quantities must be chosen to provide efficient operating conditions, and from these the required heat-transfer area of the fuel elements can be estimated. The information so obtained must then be correlated with the results of the criticality calculations and a compromise made which will provide a satisfactory balance among such economic variables as fuel inventory, reactor size, pumping power requirements, over-all thermal efficiency, etc.

12.108. While theoretical calculations can provide a general guide to core design, there are so many factors, not all of which are completely understood, to be taken into consideration that the conclusions must be subjected to experimental test. Useful preliminary information can be obtained by means of subcritical assemblies or exponential experiments, as described in § 3.191, but for the final design stages a critical assembly (or mock-up) of zero power is invaluable.* This is a flexible nuclear mock-up of the reactor core and reflector which can be rapidly rebuilt with different arrangements of fuel, moderator, etc. The critical assembly operates at virtually zero power, but the distribution of neutron flux (and of the heat generation) is essentially the same as that in the system at higher power. Some shielding for the assembly is necessary, because the neutron flux must be high enough to be measurable, but the radioactivity after shutdown is so small as to permit work being done on the assembly with little delay. Although it may not be convenient to introduce the actual coolant into a critical

* See report on speech by L. R. Hafstad in *Nucleonics*, 12, No. 10, 66 (1954).

assembly, its effects can sometimes be simulated with a solid material having almost the same scattering and absorption cross sections.

12.109. By means of such an experimental arrangement, data can be obtained concerning the effect on the critical mass, on the flux (and heat) distribution, and on the temperature coefficient of reactivity, of varying the size, design, and spacing of the fuel elements. The location of control rods and mode of operation which produces the most effective control with least distortion of the neutron flux in the reactor, and the best location for instruments, can also be studied.

Excess (Built-in) Reactivity

12.110. In addition to the bare requirements of criticality a reactor must have additional fuel included to allow for the decrease of reactivity due to such effects as increase of temperature when operating,* fuel depletion, fission-product poisoning, etc., as described in Chapter VI. For a reactor having almost pure fissionable material as fuel, there is no conversion or regeneration, so that the additional reactivity is obtained by merely summing the contributions for the various effects mentioned, as in Table 6.1.

12.111. If the reactor core contains some fertile material, so that conversion to fissionable material occurs during reactor operation, the additional reactivity is less than would otherwise be the case. Allowance has to be made for temperature and poisons, but the contribution for fuel depletion will be decreased. It will largely depend upon the conversion ratio in the given reactor and upon the expected maximum useful life of the fuel elements. If the average conversion ratio is unity (or more), during the lifetime of the fuel elements, no additional fuel is needed in the reactor to allow for depletion.

12.112. Since, however, a solid-fuel reactor must contain excess fuel at start-up, control rods must be inserted to compensate for the additional reactivity, the rods being gradually withdrawn as the fuel is consumed.† The absorption of neutrons by the control rods is wasteful, and eventually a method should be found for absorbing these neutrons in a productive manner. In addition, the presence of a strongly absorbing rod in the reactor causes a considerable distortion of neutron flux (and of the power output). This is very undesirable from the standpoint of reactor design, since it can lead to high temperatures at certain locations.

12.113. One way in which the difficulty can be partially overcome is to include a uniformly distributed ("shim") poison throughout the reactor, e.g., in the moderator or coolant. The poison captures neutrons and is thereby consumed during operation of the reactor; consequently the effectiveness of the added poison steadily decreases as the fission-product poisons accumulate and the fuel is

* Certain reactors using ordinary water as essentially nonmoderating coolant, which have the undesirable characteristic of a positive temperature coefficient of reactivity upon loss of water, are exceptional in this respect.

† If the fuel is in solution, this situation can be avoided, since additional fuel can be added as required during reactor operation.

consumed. As a result, largely automatic compensation of the decreased reactivity is provided with the minimum use of control rods.

THE HEAT-REMOVAL SYSTEM

Design Limitations

12.114. In considering the heat-removal aspect of a reactor, it is necessary to bear in mind certain limitations upon the design imposed mainly by such factors as temperature, thermal stresses, and corrosion and erosion. For example, there are definite limits set upon the temperature in the interior of the fuel element, the surface temperature of the cladding, as well as upon the temperature of the coolant. In addition, the possibility of a limitation upon the rate of flow of the coolant, e.g., due to pressure drop through the reactor or to erosion, will have an important influence on the problem of heat removal. The limitations reviewed below are applicable, in particular, to reactors with solid fuel elements and a nonboiling liquid coolant; reference to reactors of other types will be made later.

12.115. The maximum permissible interior temperature of the fuel element is determined by the possibility of phase changes. With uranium metal, for instance, it is generally considered desirable to keep the temperature below 662°C (1224°F), because of the alpha-beta phase transition (Table 8.2), accompanied by dimensional changes, which occurs at this temperature. In any case, even if this were not important, it would probably be desirable to keep the temperature below the melting point of 1133°C (2072°F). The limitation upon the temperature in the interior of the fuel element could perhaps be avoided by having the uranium in the form of a refractory compound, such as the carbide or an oxide.

12.116. The maximum surface temperature of the fuel-element cladding depends upon the nature of both the cladding material and the coolant. There is always the danger of corrosion (and erosion) of the cladding by a liquid coolant and this may increase with temperature. The maximum permissible temperature is, therefore, chosen as that at which the attack is tolerable. Another requirement is that the surface temperature should be below the saturation temperature of the coolant, e.g., water, at the existing pressure, in order to prevent local boiling. The maximum surface temperature may also be related to the temperature in the interior by the allowable thermal stresses in the fuel element, especially where large temperature fluctuations may occur or brittle material is present.

12.117. The surface temperature will, of course, have some influence on the maximum coolant temperature, but, in addition, factors external to the reactor core may have to be considered in connection with the latter. The coolant temperature must be such that there will be no vapor binding of pumps, and no appreciable corrosion of vessels, piping, or other external equipment. Further,

the over-all change in temperature of the coolant between inlet and exit from the reactor may be limited by thermal stresses or distortions which may develop in the interior of the reactor, particularly during sudden changes in power level.

12.118. The flow of coolant, either water or liquid metal, at high velocity (and high temperature) is likely to cause erosion of fuel-element cladding in the reactor core, as well as of the tubes in the heat exchanger. However, it seems that, as a general rule, the flow rate of the coolant in the reactor is limited by the pressure drop, which increases according to the square of the velocity (§ 11.152). As a rough, order-of-magnitude indication it may be taken that the maximum flow rate for water at 450 to 550°F and for liquid sodium at 900 to 1000°F is about 25 to 35 ft/sec.

Choice of Coolant

12.119. Several aspects of the relationship between the properties of various coolants and reactor characteristics have been discussed in Chapter VIII, and also in previous sections of this chapter. In the present section the treatment of coolants will refer more particularly to their influence on the thermal aspects of reactor design.*

Gaseous Coolants

12.120. The most suitable gaseous coolants (for nuclear and safety reasons) appear to be helium and carbon dioxide, and neither of these reacts with metals at the temperatures likely to be encountered in a reactor system.† The design limitations upon the fuel-element surface temperature and upon the maximum temperature of the coolant, referred to above, imposed by considerations of corrosion and erosion, therefore, are less significant for gas-cooled reactors. In

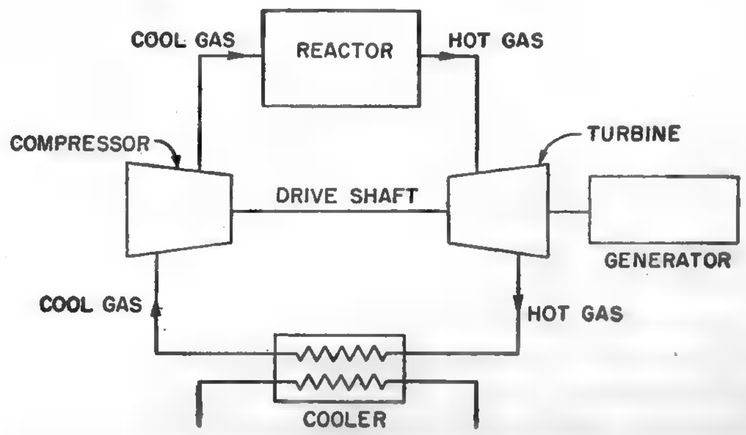


Fig. 12.1 Closed-cycle gas turbine

* Cf. F. E. Crever and T. Trocki, *Elec. Eng.*, **73**, 331 (1954).

† Carbon dioxide reacts with graphite at high temperatures, so that direct contact between these materials may have to be prevented.

addition, the problems associated with boiling of the coolant do not arise. The removal of these design limitations permits the attainment of higher temperatures than are feasible with a liquid coolant so that, in principle, higher thermal efficiency is possible.

12.121. In spite of the many advantages associated with gaseous coolants, it is felt that, largely because of their small volumetric heat capacities, such coolants are not suited to economic nuclear power production through conventional steam-electric plants. However, it is possible that a generating system, using a closed-cycle gas turbine, which lends itself to simple and compact design, might prove of interest. The basic outline of such a nuclear power plant is indicated in Fig. 12.1. The heated (compressed) gas passes from the reactor directly to a turbogenerator and then, after further cooling, to a compressor. Part of the power produced in the turbine is tapped off to compress the cooled gas before it is returned to the reactor.*

12.122. A special application of gaseous cooling of a reactor is the possible propulsion of an aircraft by means of a turbojet.† This could involve either an open-cycle gas turbine with air as the working fluid or a closed-cycle system with air as the secondary coolant. In the open-cycle system (Fig. 12.2) air is drawn in from the surroundings and is forced by the compressor through the

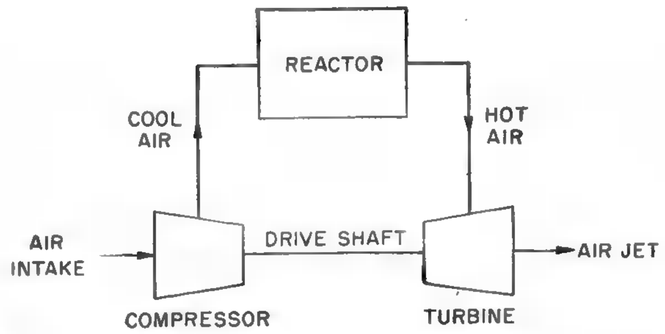


Fig. 12.2 Open-cycle turbojet with air as primary coolant

reactor where it is heated. The hot, compressed air then undergoes partial expansion in the turbine, to provide power for the compressor, and is expelled as a jet which may be used for propulsive purposes.

12.123. In a closed-cycle turbojet system the compressed air which produces the thrust is not heated directly by the reactor, but in a heat exchanger (Fig. 12.3). The primary coolant in the closed primary circuit can be a gas with good heat-transfer characteristics or, preferably, a suitable liquid. The

* See L. A. Ohlinger, *Nucleonics*, **6**, No. 3, 46 (1950); O. J. Woodruff, W. J. McShane, and W. J. Purcell, *ibid.*, **11**, No. 6, 33 (1953); also, E. A. Sheehan and A. O. White, Report R53GL186.

† L. A. Ohlinger, *loc. cit.*; C. Goodman (Ed.), "The Science and Engineering of Nuclear Power," Vol. II, Addison-Wesley Press, Inc., 1949, Chapter 17 (by C. Goodman).

advantage of the closed-cycle system is that the primary coolant can be chosen as the one most suitable for the reactor, and then the heat exchanger can be designed independently to provide the most efficient transfer of heat to the compressed air.

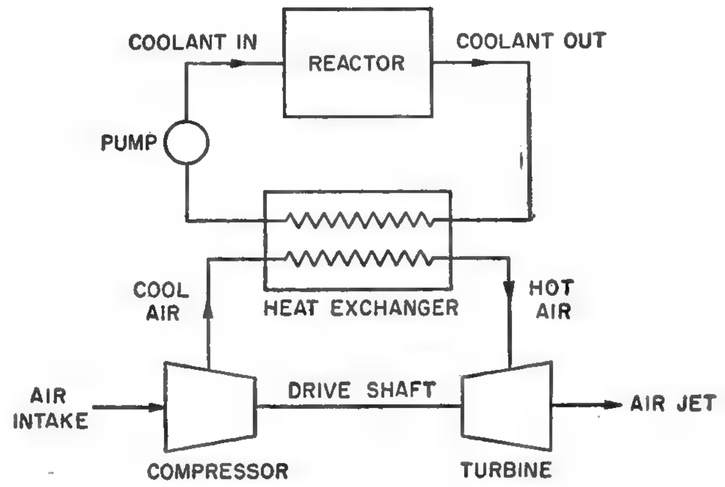


FIG. 12.3 Closed-cycle turbojet with air as secondary coolant

Liquid Coolants

12.124. It will be evident from the discussion in Chapter VIII that there is no ideal liquid, reactor coolant and, of the substances described, it is generally agreed that for thermal reactors the choice *at the present time* lies between ordinary water and liquid sodium. As seen above, these coolants are not interchangeable, for the nature of the coolant often determines the characteristics of the reactor system and vice versa (see Table 12.2).

12.125. Both water and liquid sodium have good heat-transfer properties so that there is relatively little to choose between them in this respect. The main advantage of water is its low cost, availability, and familiar technology; on the other hand, in order to attain temperatures, e.g., 500°F, at which reasonable thermal efficiencies can be realized, pressures of at least 1000 psi must be used. Further, the corrosive character of liquid water increases with temperature.

12.126. As sodium has a very high boiling point (1620°F), it can be circulated through the reactor at moderate pressures at temperatures of 1000°F or more. This is offset by the special handling equipment necessitated by the chemical reactivity of sodium with both air and water, and by its moderately high melting point (208°F). It must also be remembered that, as a result of passage through the reactor, considerable radioactivity is induced in the sodium, so that extensive shielding or a secondary cooling system is necessary. Water also acquires some activity, primarily due to the formation of nitrogen-16, but this has a half-life of only 7.3 sec, so that a short period of hold-up will permit extensive decay.

12.127. In Table 12.3 there is given a comparison of the thermal characteristics of nuclear power plants using water and sodium cooling, respectively.* Although there will inevitably be some variations, the data presented may be regarded as being fairly typical. It is expected that, owing to the higher thermal efficiency possible with sodium as coolant, the cost per kw of electrical capacity of the non-nuclear portion of the plant, i.e., the steam boiler and turbogenerator, will be somewhat less than when water is used. In both cases, however, the cost may exceed that of modern conventional power plants, because the latter generally have more favorable steam conditions at the turbine throttle.

TABLE 12.3. THERMAL CHARACTERISTICS OF TYPICAL NUCLEAR POWER PLANTS

Conditions	*Coolant*	
	Water	*Sodium*
Inlet temperature, °F	400	600
Outlet temperature, °F	500	900
Pressure, psia	1000	50–80
Turbine throttle:		
Steam temperature, °F	370	750
Steam pressure, psia	175	485
Thermal efficiency, %	24	30
Flow rate, lb/hr per kw	200	100
Non-nuclear plant, estimated cost, $/kw	130	110

12.128. To compensate for the expected lower cost of the power-generating system with a sodium-cooled reactor, allowance must be made for the cost of the special precautions which are required. The system must be leakproof and all surfaces must be blanketed with an inert gas to prevent fires. If, in spite of all precautions, access of oxygen cannot be completely prevented, an elaborate purification system is necessary to remove sodium oxide and other impurities. In addition, pipes, tanks, etc., must have heaters to prevent the liquid metal from solidifying when the reactor is shut down, and adequate shielding must be provided. Against this, allowance must be made for the cost of the pressure vessels required for a water-cooled reactor.†

12.129. It will be evident from the foregoing discussion why there are no overwhelming arguments in favor of either water or sodium as coolant for a thermal reactor. With the experience that will be gained from reactor experiments during the next few years, a clear choice may become possible. In this connection, it should be noted that if boiling-water reactors prove to be successful they will provide a third contender for power production (see § 12.149).

* J. A. Lane, *Nucleonics*, **12**, No. 6, 12 (1954).

† For reviews of engineering aspects of liquid-metal heat transfer, see T. Trocki, *et al.*, *Nucleonics*, **10**, No. 1, 28 (1952); *Mech. Eng.*, **75**, 472 (1953); Report AECU-2755; also, R. D. Brooks and A. L. Rosenblatt, *Mech. Eng.*, **75**, 363 (1953).

Method of Heat Removal

12.130. There are three general methods for removing heat from a reactor to be utilized for power purposes;* these are (1) use of an internal coolant, i.e., the coolant passes through the reactor, (2) circulating liquid fuel cooled externally, i.e., outside the reactor, and (3) boiling liquid, e.g., water, in the reactor. The selection of the method of heat removal is bound up, to some extent, with the choice of heterogeneous or homogeneous arrangement of fuel and moderator. External cooling of a circulating liquid fuel, for example, is possible only for a homogeneous fluid system,† although the other forms of heat removal can be adapted to either heterogeneous or homogeneous reactors. Unless heavy water is employed, the only liquid now considered for a boiling reactor is water and this means that the fuel must be enriched uranium. The same would be true for a homogeneous reactor with water as the fluid medium. Considerations such as these, must, of course, be borne in mind, when selecting the heat-removal procedure.

12.131. Internal cooling of the core by passage of a coolant through it has been used in nearly all reactors so far constructed or under construction. Actually, two types of internal cooling may be distinguished: (1) the coolant, e.g., air or other gas (X-10, BNL, Windscale, Saclay reactors), water (NRX, Hanford, Water Boiler reactors), or liquid metal (EBR, SIR, SRE), is not a required nuclear component; (2) the coolant is identical with the liquid moderator, which may be either ordinary water (STR, MTR, PWR) or heavy water (CP-3 and similar reactors, CP-3′, CP-5). The latter method of internal cooling is to be preferred, because, in the first place, construction is simplified; and second, since it does not involve the introduction of extraneous material, it is advantageous as far as neutron economy is concerned. However, it requires an increase in the quantity of moderator, part of which is always outside the reactor, and this will be economically significant if heavy water is used.

Circulating Fuels: Aqueous Solutions

12.132. The circulating (homogeneous) fuel reactor, with external cooling, appears to be attractive from other viewpoints in addition to that of heat removal. In internally cooled reactors, there is often a conflict between the heat-removal requirements and the optimum criticality conditions, as explained earlier. With external cooling of the fuel solution, no such difficulty arises, since the heat-removal and the nuclear systems are essentially independent. There is now no necessity for compromise and the external heat exchanger is made as efficient as possible, regardless of other circumstances. The power output of the reactor is determined by the rate of heat transfer in the heat exchanger, so

* Cooling by natural (free) convection is not included, since it could be used for power reactors only in special cases of boiling systems or for low power outputs.

† The moderator may be the fluid medium, as in the HRE, or it may be a solid, e.g., graphite, as in the LMFR (see § 12.141).

that it is possible to design simple and compact reactors of high specific power. Further, there is less restriction upon the choice of secondary coolant; as it does not enter the reactor, a material which absorbs neutrons readily can be used if its thermal properties are desirable. As a general rule, the actual coolant will be the water in a steam generator in which heat is removed from the fuel solution.

12.133. Among other advantages of a circulating homogeneous fuel are elimination of radiation damage to fuel elements and of the accompanying limitation upon fuel burn-up. There is some decomposition of water by radiation, but this does not seriously affect operation of the reactor at sufficiently high temperatures and pressures (§ 8.141). Before the HRE was operated it was feared that nuclear instability might result from the liberation of hydrogen and oxygen gas bubbles in the fuel solution, but this has been found not to be the case at the power densities attained hitherto.

12.134. Processing of the fuel is simplified, since the uranium is already in solution, and metallurgical operations and fabrication of fuel elements are unnecessary. By withdrawing quantities of fuel solution from time to time for removal of fission products (and of plutonium-239 or uranium-233, if required) and replacing it with fresh material, essentially continuous operation is possible, without the need for shutting down to discharge (unload) and recharge (load) the reactor. Additional fuel can also be added to compensate for that consumed during operation.

12.135. One of the outstanding attributes of a homogeneous fuel reactor, especially when either ordinary or heavy water is the liquid medium, is the ease of control. The change in density of the solution with temperature leads to a sensitive negative temperature coefficient, which makes the reactor self-stabilizing. The HRE, for example, has been found to function satisfactorily without the use of regulating rods (§ 13.102). Any increase in external power demand is accompanied by a decrease in the temperature of the fuel solution returning to the reactor. There is then a very rapid response in an increase of reactivity. As the heat-release rate in the reactor approaches the external demand the reactivity falls until the effective multiplication factor is unity and the power output is steady. Shim control rods are also unnecessary, for the reactor can be started up and shut down, gradually, by changing the concentration of the fuel solution. It was thought at one time that circulating-fuel reactors might prove difficult to control because some of the delayed neutrons are emitted outside the reactor, but this fear has proved groundless.

12.136. As with many other aspects of reactor design, there are drawbacks to weigh against the advantages of circulating-fuel reactors. Perhaps the most outstanding of these are the possibility of leakage of the fuel solution and the problems of corrosion and erosion. Some aqueous solutions of uranium salts, especially at high temperature, readily attack many metals that would normally be used for pipes, pumps, heat exchangers, etc. The fact that the solutions

become highly radioactive during reactor operation and are under high pressure adds greatly to the operational hazards. The fuel solution leaving the reactor is no longer critical, but it continues to emit delayed neutrons, in addition to beta and gamma radiation, so that radioactivity may be induced in the heat exchanger and coolant. Adequate shielding of the heat exchanger is necessary and maintenance requires special care.

12.137. The decomposition of ordinary or heavy water by radiation will produce a considerable quantity of hydrogen (or deuterium) and oxygen gases during operation of the reactor, namely, about 10 ft^3/min (at S.T.P.) per megawatt of reactor power (§ 8.184). Apart from the danger of explosion of the gas mixture, against which special precautions must be taken, it is possible that the gas bubbles may cause instability of the reactor at high power densities, although, as stated above, no such instability has been observed during operation of the HRE.

12.138. The HRE, which is described more fully in § 13.98, *et seq.*, was designed as an experiment on a moderate scale (1000 kw) to determine whether the advantages of a circulating-fuel reactor could be realized and the drawbacks overcome. The results have been so encouraging that HRE-2 (or HRT), which is to have a heat output of 5000 kw, is to form part of the A.E.C.'s Five-Year Program. The primary purpose of this experiment is to develop a simple, mechanically satisfactory plant which will demonstrate the operability and reliability of a reactor having a circulating aqueous fuel solution under conditions simulating full-scale operation. The HRE-2 (or HRT) is to be followed by a further step-up in power to the 65,000-kw output of the Homogeneous Thorium (Breeder) Reactor (HTR). Both of these reactors will have associated chemical plants for processing of the fuel solutions, thus permitting an estimate to be made of the over-all cost of power production.

Aqueous Slurries as Circulating Fuel

12.139. An interesting possibility for obtaining a fluid fuel system that has a high concentration of uranium yet is not so corrosive is to use a slurry or suspension of an insoluble compound, such as an oxide, in either ordinary or heavy water. Although a uniform slurry is heterogeneous in the strict physical sense, it will behave as a homogeneous fuel in a reactor. Consequently, if ordinary water is the medium, an enriched fuel would be essential; with heavy water, however, uranium of normal isotopic composition would be adequate, although some enrichment might be beneficial.

12.140. Among the predicted advantages of a slurry are greater chemical and radiation stability than a solution and, possibly, a decrease in corrosive action. On the other hand, if the suspended particles are abrasive, they may cause mechanical wear, i.e., erosion, in a dynamic system. The chief question arising in connection with the feasibility of using a slurry as circulating fuel for a reactor is the maintenance of the suspension in a uniform state by preventing settling or

coagulation of the suspended particles, especially when the circulation is stopped. There are various ways in which this may be achieved. For example, the settling of suspensions of uranium oxides in water may be inhibited (or prevented) if the medium is made slightly alkaline, i.e., if the pH exceeds 7.* Alternatively, since the particles will probably remain suspended while the slurry is being circulated, all that it is really necessary is to be able to redisperse them, e.g., mechanically, if they have settled out while the reactor is shut down.

Nonaqueous Fluid Fuels

12.141. In order to obtain reasonable thermal efficiency, a reactor using an aqueous slurry as fuel must, like other power reactors containing ordinary or heavy water, be operated at high pressures. An extensive study has therefore been made of a graphite-moderated Liquid Metal Fuel Reactor (LMFR) which can be adapted to either internal or external cooling.† The medium is liquid bismuth which dissolves uranium metal to the extent of about 0.5 per cent at 500°C (932°F); this is regarded as being more than ample for the attainment of criticality in an enriched-fuel reactor with graphite as moderator. The solubility of uranium in bismuth increases with temperature, so that, if suitable constructional and containing materials are available, a reactor of high specific power and excellent over-all efficiency should be attainable. Some of the difficulties associated with the use of liquid bismuth were mentioned in § 8.246, and it was indicated there how they may be overcome.

12.142. Because of the relatively low solubility of elemental uranium in liquid bismuth, the possibility has been considered of using suspensions of solid uranium-bismuth or uranium-tin compounds in either liquid bismuth or in mixtures with lead and tin. The latter two elements were selected partly on account of their small cross sections for neutron absorption. It has been found that the solid compound of uranium and tin, USn_3, has almost the same density (10 g/cm^3) as a low melting-point (240°F) alloy of bismuth (56% by weight), lead (38%), and tin (6%). A suspension of solid USn_3 in this liquid would be expected to show little tendency to settle.

12.143. As part of the over-all design of the LMFR, it has been suggested that the reactor might be used as a uranium-233 breeder. The blanket would consist of the graphite reflector pierced by tubes containing thorium in the form of the compound Th_3Bi_5 suspended in liquid bismuth (or in a molten bismuth-lead alloy) which has almost the same density. The heat generated in the blanket could be removed externally by circulation of the fluid.

12.144. The designers of the LMFR regard its most important feature as integration of the reactor system with chemical processing of the fuel. The

* Cf. J. J. Went and H. de Bruyn, *Chem. Eng. Prog. Symposium Series*, No. 12, **50**, 120 (1954); *Nucleonics*, **12**, No. 9, 16 (1954). In these papers a design for a reactor using a circulating, slurry-type fuel is outlined; there are, however, several unpublished reports of earlier date concerning such reactors and the properties of reactor-fuel suspensions.

† See references to § 8.240.

removal of fission products from the spent fuel solution by the salt-extraction method was described in § 7.196. The integration of reactor design with the recovery of the fuel is highly desirable if maximum over-all economy is to be achieved.

12.145. As an alternative to liquid-metal fuels for the attainment of high temperatures, without the necessity for excessive pressures, the use of fused salt or hydroxide systems has been suggested. An ideal system of this type should have a fairly low melting point and also a low vapor pressure. It should be stable to both temperature and radiation and should be capable of holding appreciable amounts of fuel material in solution (or suspension). Finally, the elements present should have small capture cross sections for neutrons and, if the liquid fuel is to be used in a homogeneous thermal reactor, it is desirable that the elements, other than uranium (or plutonium), present should be effective as moderators, i.e., they should have low mass numbers.

12.146. The foregoing requirements are met to a large extent by mixtures of fluorides (see § 8.253) which can dissolve one or other of the fluorides of uranium. Several such mixtures have melting points which are not excessively high, e.g., 400°C (750°F), although they are well above normal temperatures. A possible advantage of the use of a fluoride fuel system is that it might perhaps be combined with the volatilization method of separating uranium (as hexafluoride) from fission products and other elements (§ 7.200).

12.147. The alkali metal hydroxides, e.g., those of lithium, sodium, and potassium, have fairly low melting points and are thermally stable, although little is yet known of their stability to the radiations to which a reactor fuel is exposed. Fused hydroxides of lithium-7 and, perhaps, sodium (or mixtures of these two) might form a basis for a thermal reactor fuel system (§ 8.254). It would probably be advantageous to employ deuteroxides rather than compounds containing normal hydrogen in some possible applications.

12.148. Unfortunately, the solubility of uranium in fused alkali hydroxides is relatively low except at such high temperatures that finding suitable containing material becomes a serious problem. However, as indicated above, it is not essential that the uranium be in solution. It is possible, therefore, that suspensions of uranium oxide in fused hydroxides (or deuteroxides) may provide feasible liquid reactor-fuel systems. Such systems could presumably be employed at temperatures lower than those necessary to produce true solutions of adequate concentration.

Boiling-Water Reactors

12.149. The last method of reactor cooling to be reviewed here is that involving the boiling of water (or heavy water) and the production of steam in the reactor core, as outlined in § 8.238.* From the design standpoint, boiling-water reactors have numerous advantages. Since steam is generated directly

* For references, see § 8.239.

in the reactor, an external steam generator (heat exchanger) is not required. As a result, in addition to a considerable financial saving, the same steam temperature can be attained with a lower average fuel temperature, or a higher steam temperature is possible for a specified fuel temperature. Because no excess pressure is required to prevent boiling of the water in the reactor, as is necessary in water-moderated power reactors of other types, the pressure vessel can be designed for a lower pressure. In the PWR, for example, the pressure in the vessel containing the core will be 2000 psia, but the pressure of saturated steam leaving the steam drum is expected to be only 600 psia. In a boiling-water

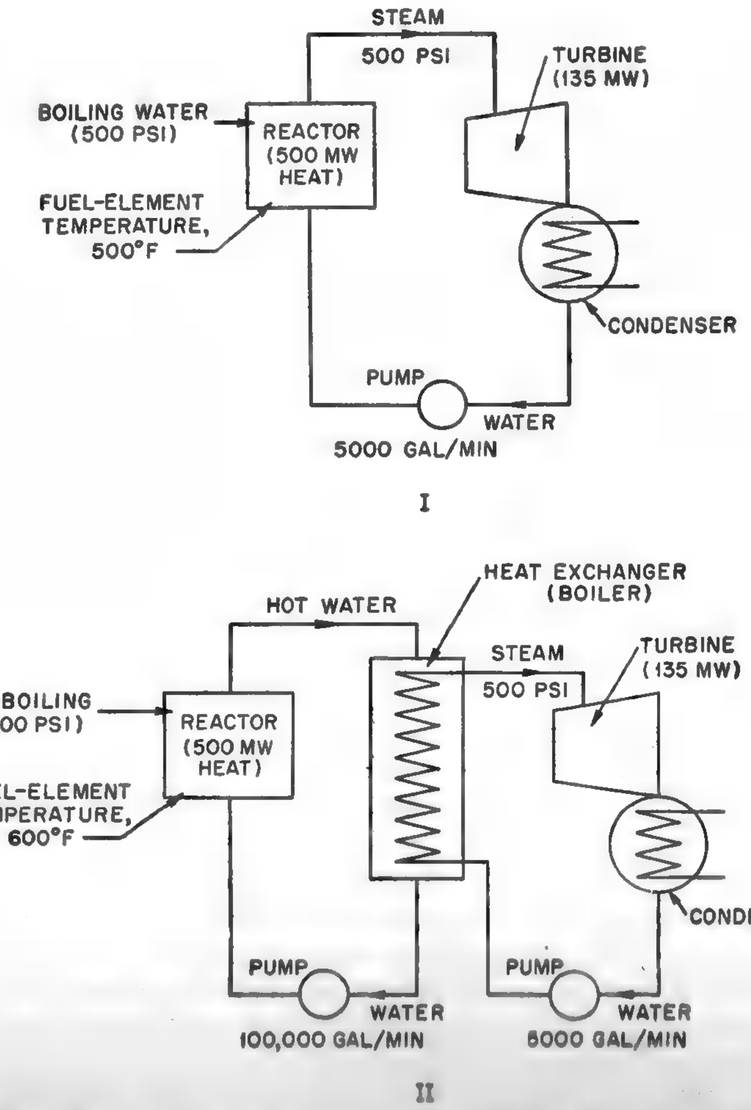


Fig. 12.4 Comparison of (I) boiling-water and (II) water-cooled reactors. (S. Untermyer)

reactor, the same steam pressure could be achieved with a reactor-vessel pressure of little more than 600 psia.

12.150. The pumping requirements for a boiling-water reactor are only those needed for returning water from the turbine condenser to the reactor vessel. It is estimated that pumping power will be about one tenth of that for a system having a separate steam generator. A rough comparison between the conditions of operation of a boiling-water reactor power-plant and one using pressurized-water cooling is shown in Fig. 12.4.*

12.151. The coolant, in the form of steam, leaving the boiling-water reactor will have a lower density than liquid water, and so its radioactivity will tend to be less intense; this will mean that less shielding will be necessary for the piping. Further, because of the evaporation within the reactor, soluble radioactive material will largely remain behind in the water and not be expelled with the steam, provided a good entrainment separator is used. On the other hand, the presence of steam in the reactor core increases the neutron leakage, thereby decreasing the conversion ratio and also requiring a higher enrichment of the fuel. In addition, the specific power of a boiling reactor may be less than can be realized in a nonboiling system.

12.152. It is believed that a boiling-water reactor will be similar to a homogeneous aqueous reactor, e.g., the HRE, with regard to simplicity of control and inherent stability. However, it is not yet known with certainty that this will be the case in a reactor having a high power density and large output, suitable for commercial power production. Another possible difficulty may be the slight radioactivity of the steam entering the turbine. There will be a minor problem of preventing leakage, but a more serious matter is the question of the extent to which normal maintenance will be impeded by the deposition of radioactive material in the turbine, condenser, and feed-water pumps. The radioactivity of the steam would be much greater if a boiling aqueous solution were used as a reactor fuel, due to the presence of gaseous fission products. However, the present plans for a boiling-water reactor are based on the use of solid fuel elements with suitable cladding to retain the fission products (§ 13.77), although a homogeneous boiling reactor is being considered.

12.153. Since 1953 a number of experiments have been made on boiling the water moderator-coolant in a reactor with solid fuel elements. It was found that, provided the power density was not too large, the reactor would operate steadily under conditions in which steam was continuously formed and ejected from the reactor. When the reactivity was increased suddenly, the decrease in moderator density, due to the blanketing of the fuel elements with steam, resulted in a quenching of the fission-chain reaction long before the temperature became high enough to cause any damage.†

* S. Untermyer, *Nucleonics*, 12, 7, 43 (1954).

† In an experiment in July, 1954, the power of a boiling-water reactor was allowed to rise well over a million kilowatts in a tenth of a second. This reactivity excursion resulted in the destruction of the reactor core and some associated equipment. However, the conditions imposed were very much more severe than are expected to occur in any reasonably conceivable accident (Seventeenth Annual Report of the Atomic Energy Commission, U. S. Government Printing Office, January 1955, pp. 22–23).

12.154. Because of the success of the preliminary tests an experimental boiling-water reactor (EBWR), producing about 20,000 kw of heat, is being constructed to explore further the possibilities of a stable, self-regulating, power reactor which will produce steam directly. With this will be associated a turbogenerator plant, of 5000 kw electrical capacity, the purpose of which is partly to investigate the extent of radioactive contamination of the turbine system.

12.155. The stability of a solid-fuel, boiling-water reactor depends, in the main, upon three factors which change the reactivity when steam bubbles are formed. First, due to removal of the moderator, there is a decrease in the resonance escape probability, if natural or slightly enriched uranium is the fuel; this results in a decrease of the reactivity. Second, an increase in the thermal utilization, due to expulsion of the neutron-absorbing coolant and moderator, gives, on the other hand, an increase of reactivity. Third, an increase in the leakage of neutrons causes a decrease in the reactivity. In reactors having heavy water as moderator-coolant, the first and last factors always outweigh the effects of the second one. Thus steam formation tends to decrease the reactivity, giving the desired self-regulating property to the boiling reactor. If ordinary water is the moderator-coolant, however, the increase in the thermal utilization, accompanying removal of the water, is considerable, because of the significant neutron absorption cross section of hydrogen. By proper design of the reactor, so that the ratio of ordinary water to appropriately enriched uranium is kept below a certain value, the increase in reactivity due to increase of the thermal utilization, when steam is formed, can be outweighed by the decrease due to the changes in the resonance escape probability and neutron leakage. A boiling-water reactor of this type will be stable and self-regulating, although at high power densities oscillations may be expected.

Internally-cooled Homogeneous Reactor

12.156. An attractive alternative to a direct-boiling homogeneous reactor, which has a distinct possibility for a fairly small reactor unit, is a design in which water is boiled and forms steam in coolant channels passing through the homogeneous core.* Such a system would have some, but not all, of the advantages of a boiling-water reactor, on the one hand, and some of a homogeneous reactor, e.g., simplified processing of fuel, on the other hand. Since the radioactive uranium solution would not be circulated for external cooling, both the shielding and erosion problems would be decreased. If the reactor is to be stable in operation, the limitations upon the ratio of water to uranium in the reactor, mentioned above, would apply as in the case of a boiling-water reactor.

* See, for example, L. B. Borst, *Nucleonics*, 12, No. 8, 78 (1954); Atomic Industrial Forum Report, May 1954, C-2.

Fuel Element Design

12.157. If, after considering the fuel and moderator materials available and the relative merits of homogeneous and heterogeneous systems, the decision is to construct a reactor with solid fuel elements, the next problem to be considered is the design of these elements.

12.158. Ideally, a fuel element should be able to withstand high temperatures and should have good radiation stability; it should be cheap and easy to fabricate and should lend itself to simple reprocessing. In addition, it should be designed to facilitate the removal of heat produced within it. Bearing in mind the temperature and other restrictions mentioned earlier, it can be understood that the specific power attainable in a reactor will be largely dependent upon the type and design of the fuel elements.

12.159. Apart from CP-1 and CP-2 in which the fuel is (or was) in the form of lumps, the fuel elements in all the reactors using natural uranium, e.g., X-10, BNL, CP-3, BEPO, GLEEP, ZEEP, NRX, JEEP, etc., are (or were) solid rods of about an inch in diameter. The dimensions were chosen so as to take advantage of the increase in the infinite multiplication factor due to the gain in both the resonance escape probability and the fast-fission factor resulting from the heterogeneous arrangement (Chapter III). This particular design of fuel element was satisfactory for reactors of low specific power, such as those mentioned above. For a reactor of high power, however, the specific power attainable with these elements, within the existing temperature limitations, is too small to be practicable, so that changes in design are imperative.

Cylindrical Fuel Elements

12.160. With the availability of enriched uranium fuel, some sacrifice is permissible in the infinite multiplication factor for the sake of the improvement in heat-transfer characteristics of the fuel elements. By decreasing the diameter of the uranium rods, it is possible to achieve a considerable increase in the specific power of the reactor for a given temperature drop between the uranium interior (t_0) and the coolant (t_f). Disregarding the cladding, the effect of which is not significant, the temperature difference, for an unclad cylinder of radius a, is obtained from equation (11.87.2) by setting $a = b$; thus,

$$t_0 - t_f = \frac{Qa^2}{4k} + \frac{Qa}{2h}$$

or

$$Q = \frac{4hk(t_0 - t_f)}{a(ah + 2k)}, \qquad (12.160.1)$$

where Q, the heat source per unit volume, is directly related to the specific power of the reactor. It is immediately evident from equation (12.160.1) that for a given value of $t_0 - t_f$ the specific power will increase with decreasing radius of the cylindrical fuel rod.

Example Compare the maximum specific power possible for a water-cooled reactor with cylindrical rods of uranium 1 in. in diameter with one in which the rods are 0.5 in. in diameter, for a given fuel-coolant temperature drop. For uranium k is 14 Btu/(hr)(ft²)(°F/ft) and h may be taken as 5000 Btu/(hr)(ft²)(°F) in each case.

Let Q_1 be the volumetric heat source for a rod of diameter 1 in., i.e., radius 0.5 in., and let $Q_{0.5}$ be the value for the rod of diameter 0.5 in., i.e., radius 0.25 in. Then, from equation (12.160.1), since the numerator on the right is constant,

$$\frac{Q_{0.5}}{Q_1} = \frac{0.5[(0.5/12)(5000) + (2)(14)]}{0.25[(0.25/12)(5000) + (2)(14)]}$$
$$= 3.6.$$

The specific power attainable with the 0.5 in. diameter rods is thus 3.6 times as great as that with the 1 in. rods.

12.161. By reducing the diameter of a cylindrical fuel element, the specific power attainable for given temperature drop can be increased or, alternatively, a given specific power can be obtained with a smaller temperature drop and its consequent advantages, e.g., decrease in thermal stresses within the elements. However, it must be remembered that because of the necessity for cladding the fuel, as explained below, a decrease in the fuel-rod radius means an increase in the amount of cladding material for a given mass of fuel. The accompanying increase in parasitic neutron capture may seriously affect the over-all neutron economy of the reactor. Further, the use of thin rods as fuel elements will, undoubtedly, increase the total cost of fabrication. If cylindrical elements are to be employed, the diameter chosen will represent a compromise between specific power, on the one hand, and both neutron economy and cost on the other hand. A diameter of 0.25 in. is probably close to the optimum value. It may be mentioned that by arranging a number of thin rods in groups so that they are less than a neutron mean free path apart (Fig. 12.5), much of the advantage of a thick rod, in respect to resonance escape and fast-fission factors can be retained.

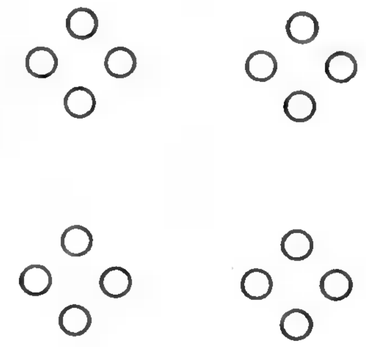


Fig. 12.5 Possible grouping of thin fuel rods

12.162. The use of a moderately thick, cylindrical fuel element with a central channel, so that it can be cooled both internally and externally, has been proposed. In a sense this is almost equivalent to the close grouping of thin rods and has advantages from both nuclear and heat-transfer points of view. However, in the past such elements were not only difficult to fabricate, but have been found to be unsatisfactory because of the development of a high thermal resistance at the inner fuel-cladding interface due to thermal expansion effects.

Plate-type Fuel Elements

12.163. An alternative possibility, which has much to recommend it, for water-moderated reactors, is the plate design for fuel elements; it has been used

in several of the more recent research reactors, e.g., MTR, BSR, CP-5. The fuel element consists of a set of long slightly curved (or flat) plates of a uranium-aluminum alloy with a thin cladding of aluminum on both sides, resulting in a sandwich-type of construction. The plates are held in position in a long box-like frame, forming a composite fuel element. A horizontal section through such an element, containing six clad plates, is shown in Fig. 12.6 (see also Fig. 13.13).

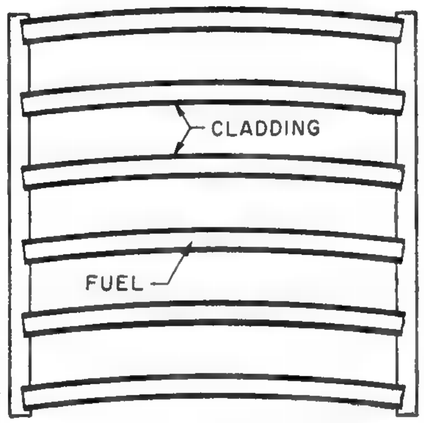


FIG. 12.6 Assembly of sandwich-plate type fuel element

12.164. The sandwich-plate type of fuel element is relatively simple to fabricate,* and the parasitic capture of neutrons can be kept relatively small by using a thin cladding, just sufficient to retain the fission products and protect the uranium from corrosion. If the fuel is natural (or slightly enriched) uranium, it is possible, by proper spacing of the fuel plates, e.g., by placing them close together and decreasing the moderator-to-fuel ratio, to obtain an appreciable gain in the fast-fission factor accompanied by a decrease in the resonance escape probability. If the fuel-element boxes are closely spaced, as is frequently the case, the system may be treated as homogeneous for theoretical analysis (§ 12.105).

12.165. In addition, high specific powers are possible in reactors using plate-type fuel elements. For a large unclad fuel plate, the temperature drop between the fuel and the coolant is obtained from equation (11.85.2) by setting a equal to b; the result is

$$t_0 - t_f = \frac{Qa^2}{2k} + \frac{Qa}{h}, \tag{12.165.1}$$

where a is the half-thickness of the fuel region. From this expression, which is analogous to that given above for a cylindrical fuel rod, it is possible to calculate the specific power attainable in the reactor for a given temperature drop.

Example Suppose the average difference in temperature between the interior of the fuel plates and the coolant in a reactor is 200°F. What will be the specific power of the reactor, having fuel plates 0.25 in. thick, consisting of uranium metal and containing 1.2 per cent of uranium-235? For uranium, k is 14 Btu/(hr)(ft²)(°F/ft) and h may be taken as 5000 Btu/(hr)(ft²)(°F).

From equation (12.165.1)

$$Q = \frac{2hk(t_0 - t_f)}{a(ah + 2k)}$$

* With increasing experience fabrication costs can be reduced; thus, the cost of fabricating the fuel elements for the MTR was decreased from about 65 cents/million Btu of heat in August, 1952 to 10 cents/million Btu in July 1953 (L. H. Hafstad, Atomic Industrial Forum Report, July 1954, p. 31).

and, since a is $(\frac{1}{2})(0.25)$ in. = 0.25/24 ft, it follows that

$$Q = \frac{(2)(5000)(14)(200)}{(0.25/24)[(0.25/24)(5000) + (2)(14)]}$$
$$= 3.4 \times 10^7 \text{ Btu/(hr)(ft}^3\text{)}.$$

To convert Btu/hr to kw, it is necessary to multiply by 2.93×10^{-4}; hence, Q is $(3.4 \times 10^7)(2.93 \times 10^{-4}) = 1.0 \times 10^4$ kw/ft³. The density of uranium is 19 g/cm³, so that 1 ft³ weighs 538 kg; since it has been postulated that the uranium contains 1.2 per cent of uranium-235, it follows that 1 ft³ fuel contains (538)(1.2)/100 = 6.46 kg of uranium-235. The specific power of the reactor, which is equivalent to Q, in the appropriate units, is then

$$Q = \frac{10^4}{6.46} = 1530 \text{ kw/kg of uranium-235}$$
$$= \frac{10^4}{538} = 18.6 \text{ kw/kg of uranium.}$$

12.166. The data in the foregoing example are not at all unreasonable; in fact, they may be somewhat conservative for a power reactor. The average temperature drop of 200°F between fuel and coolant allows for a maximum of at least 500°F; hence a temperature as high as 500°F for the coolant means that the temperature of the fuel interior will not exceed 1000°F. The suggested plate thickness of 0.25 in. is by no means as small as can be fabricated, and the use of a uranium alloy, instead of pure uranium, as in the example, would serve to increase the specific power. Finally, an increase in the heat-transfer coefficient, which can be readily achieved, would also result in an increase in the specific power. It is seen, therefore, that specific powers as high as (or higher than) 24 kw/kg of uranium should be readily attainable in a reactor with plate-type fuel elements of slightly enriched material.

CLADDING

12.167. Cladding of reactor fuel is necessary for several reasons; the two most important are (1) to prevent attack of the uranium, thorium, or plutonium by air, water, or other coolant, especially at high temperatures, and (2) to prevent escape of the radioactive fission products, including gases, and of plutonium, which is a serious health hazard (§ 9.74). Aluminum has been extensively used as cladding material; less than 0.001 in. (1 mil) is sufficient to stop fission products, although a greater thickness is desirable for mechanical strength, to prevent penetration of corrosion pits, and to retain the gases. Because it is itself attacked by water at elevated temperatures, the limiting acceptable surface temperature, with water as coolant, is about 400°F. With gaseous coolants, e.g., helium or carbon dioxide, higher temperatures might be permissible, although the rapid loss of strength suffered by aluminum above 400°F would be a limitation.

12.168. Due to its satisfactory nuclear and mechanical properties, and its resistance to corrosion, zirconium is attracting interest for fuel cladding in spite

of its high price (§ 8.191, *et seq.*). The maximum permissible surface temperature in water is near 660°F, although this might well be extended by alloying with other metals. Maximum water temperatures of about 560°F are thus possible with zirconium (or, better, zirconium alloy) cladding; this is probably as high as is practical at the present time in nonboiling reactors because of the pressure required to prevent boiling, i.e., 2000 psia.*

12.169. Neither aluminum nor zirconium would be satisfactory fuel-cladding materials for a sodium-cooled reactor, if advantage is to be taken of the high temperatures which can be attained. In this case, stainless steel appears to be the only reasonable possibility at the present time. Its surface temperature can be permitted to rise to 1200°F, and it is resistant to attack by liquid sodium. Although it captures neutrons more readily than zirconium, stainless steel is much cheaper, and so it has been suggested as an alternative cladding material for water-cooled reactors.† Some consideration has been given to titanium (or its alloys) for sodium-cooled reactors, in spite of its high capture cross section for thermal neutrons (5.6 barns).

12.170. Another aspect of cladding is that, if the material has suitable mechanical properties, the clad fuel will be more able to withstand thermal stresses due to temperature variations, and dimensional changes due to radiation damage. In these respects it is probable that plate-type fuel elements, with thin fuel layers, are superior to cylindrical elements.

12.171. In choosing the cladding material, the problem of its removal, as the first stage in processing the spent fuel, must be borne in mind. Aluminum can be dissolved away readily, but zirconium and stainless steel are more difficult to remove. The latter can be dissolved fairly easily if it is first carburized. If the cladding could be detached mechanically, the situation might be greatly simplified. This appears to be a possible development in fuel-element design.

12.172. In order to keep thermal resistance to a minimum, it is desirable that there should be a good contact (or bond) between the fuel and the cladding. There are several ways of achieving satisfactory bonding. With plate-type fuel elements, bonding may be achieved by hot-rolling the fuel-cladding sandwich. For cylindrical elements, a liquid metal, such as sodium or NaK, can be included before the "can" of cladding is sealed. It is reported that aluminum cladding can be bonded to the fuel by means of an alloy of aluminum and silicon.

* The boiling point of water at 2000 psia is about 635°F, but it is the practice to allow a differential of at least 40°F to avoid the possibility of local boiling (cf. § 12.116).

† In view of the fact that neutrons captured in stainless steel would otherwise be available for the production of fissionable nuclei, it is a moot point whether this material is really more economical than zirconium for water-cooled reactors (see W. E. Abbott, *Chem. Eng. Prog.*, **50**, 245 (1954)).

Reactor Coolant Temperatures*

12.173. The selection of the inlet and outlet temperatures for the reactor coolant is a complex matter having economic as well as engineering and thermodynamic aspects. With the exception of a few special cases, generation of electric power from the heat liberated in a reactor will be based on the production of steam in a heat exchanger (or boiler), the steam being utilized in a turbogenerator. In choosing the operating temperatures for the reactor coolant it is necessary to take into consideration the conditions in the steam-turbine system as well as those in the reactor and their mutual interaction.

12.174. Unless some additional form of heating is used, which is assumed not to be the case, the temperature of the steam produced in the boiler (with or without superheating) cannot exceed the temperature at which coolant leaves the reactor. Since the thermal efficiency of the turbine increases with steam temperature it would appear that the coolant exit temperature from the reactor should be as high as possible in order to permit the attainment of high steam temperatures.

12.175. The highest steam temperatures used in conventional central-station power plants now approach 1200°F, but values of about 1050°F are more common.† This limit is set, to a large extent, by the increasing cost per electrical kw of the special materials required to withstand high temperatures. From the opposing effects of the more expensive equipment and the increase in thermal efficiency, it has been concluded that there is an optimum temperature for the steam at the turbine throttle in conventional power plants. It is possible that a somewhat similar conclusion may apply to nuclear-power plants, but this is by no means certain. In any event, because of the difference in fuel costs, the optimum steam temperature may be lower in a nuclear plant.‡

Coolant Outlet Temperature

12.176. The maximum possible exit temperature of the coolant from the reactor is determined by economic factors, e.g., the increasing cost of the pressure vessel with operating temperature, if water is the coolant, as well as by physical considerations, e.g., the maximum permissible temperature of the interior of the fuel or of the surface of the cladding in contact with the coolant. If one of these temperatures is limiting, then the maximum possible rate of heat removal from the reactor, i.e., its maximum possible operating power,§ for a given coolant flow rate can be represented, to a good approximation, by the expression

* A. Amorosi, Argonne National Laboratory, unpublished lectures, 1949; W. E. Parkins, *Conf. on Nucl. Eng.*, Berkeley, 1953, F-55; W. A. Loeb, *Chem. Eng. Prog. Symposium Series*, No. 11, **50**, 145 (1954); F. T. Miles and I. Kaplan, *ibid.*, No. 11, **50**, 159 (1954); C. H. Robbins, *ibid.*, No. 12, **50**, 181 (1954).

† These are the temperatures in plants where fuel costs are relatively high, i.e., 30 to 50 cents/million Btu.

‡ See R. D. Elliott, *Nucleonics*, **10**, No. 2, 57 (1952), **10**, No. 8, 82 (1954); T. Trocki and J. D. Selby, *ibid.*, **10**, No. 8, 82 (1954).

§ The actual maximum operating power is smaller, for reasons which will be apparent shortly

$$q_{\max} \approx A_h H(t_{\lim} - t_{\text{out}}), \qquad (12.176.1)$$

where $t_{\lim}$ is the limiting temperature and t_{out} is the average coolant temperature at the reactor outlet; A_h is the heat-transfer surface in the reactor, and H is an effective coefficient of heat transfer. It follows, therefore, from equation (12.176.1) that if A_h and H are specified, the maximum reactor power will decrease with increasing coolant outlet temperature, since $t_{\lim}$ may be regarded as being fixed.

12.177. As seen above, the maximum thermal efficiency of the turbine can be increased as t_{out} is increased, because of the higher possible steam temperature. But if the maximum reactor power decreases at the same time, it follows that there will be an optimum coolant outlet temperature which will give a maximum electrical power output. It should be noted, however, that, although $t_{\lim}$ is fixed, the power output for a given value of t_{out} can perhaps be improved by increasing A_h and H by suitable design changes. For example, an increase in the flow rate, if permitted by the pressure drop, will be accompanied by an increase in the heat-transfer coefficient of the fluid. This will be reflected in some increase of H, the effective (over-all) coefficient of heat transfer.

12.178. In addition to the optimization of the coolant outlet temperature on the basis of heat-transfer considerations within the reactor, there may be an optimum determined by conditions in the boiler, as will be seen shortly. Although a lower steam temperature means a decrease in the turbine thermal efficiency, there will be a compensatory saving in the size and cost of the boiler. Consequently a decrease in the steam temperature, since it can be accompanied by a decrease in the coolant outlet temperature, and a decrease in the pressure within the reactor if water is the coolant, may sometimes have an over-all economic advantage.

Coolant Inlet Temperature

12.179. Suppose that, after giving due consideration to the various factors reviewed above, a tentative decision has been reached concerning the average temperature at which the coolant is to leave the reactor. A choice must now be made of the coolant inlet temperature, and here also various factors must be balanced against one another. According to equation (11.137.2) the rate of heat removal from a reactor is given by

$$\begin{aligned} q &= (\rho c_p) v A_f \Delta t_c \\ &= (\rho c_p) v A_f (t_{\text{out}} - t_{\text{in}}), \end{aligned} \qquad (12.179.1)$$

where ρc_p is the volumetric heat capacity of the coolant, v is its flow rate through the reactor, A_f is the total flow area, and Δt_c is the difference between the temperatures at which coolant leaves (t_{out}) and enters (t_{in}) the reactor.

12.180. Since ρc_p is fixed by the nature of the coolant and t_{out} may be regarded as more or less determined, as described above, it is seen that the value of t_{in} will depend upon q, the total heat power output of the reactor, as well as upon the coolant velocity and the flow area. From a knowledge of the expected power level, calculations involving nuclear and heat-transfer considerations are made, based on tentative decisions concerning fuel-element design and coolant-passage geometry (see § 12.193, *et seq.*). These give a provisional value of A_f, the coolant flow area, and the related equivalent diameter of the channels.

12.181. The next step is to estimate the available pressure drop or pumping power for the coolant. This is largely determined by economic factors, such as cost of pumps and piping, the size of the heat exchanger, and the value of the coolant (or of the fuel in a circulating liquid-fuel system). In this connection a preliminary plant layout of the reactor and its primary cooling system, and a knowledge of heat-exchanger designs for a wide variety of conditions, will be necessary.

12.182. When rough values of the pumping power and flow area, which appear to be reasonable, are available, it is possible to establish the maximum coolant flow velocity, assuming that, as is generally the case, the pressure drop is the limiting factor. A large value of v is desirable, if conditions permit, since it is accompanied by an increase in the heat-transfer coefficient. However, beyond a certain point the gain may not be worth while, e.g., with a liquid-metal coolant (§ 11.133), because of the increased cost of pumping. With q, ρ, c_p, v, A_f, and t_{out} being, at least provisionally, known, the corresponding value of t_{in}, the coolant inlet temperature to the reactor, can be obtained from equation (12.179.1).

12.183. It is of interest to call attention to the influence of the volumetric heat capacity of the coolant on t_{in} or, actually, on Δt_c, the over-all increase in temperature of the coolant. For water, in the useful temperature range of 350° to 450°F, the value of ρc_p is about 56 Btu/(ft³)(°F), whereas for liquid sodium, in the range from 700° to 1000°F, it is roughly 16 Btu/(ft³)(°F). It is seen, therefore, from equation (12.179.1), that if v and A_f are not very different for two reactors using water and sodium, respectively, as coolants then, in order to remove heat at the same rate, Δt_c will have to be at least three times as great for the sodium reactor as for the one with water as the coolant (cf. Table 12.3). A reactor using liquid sodium as coolant should therefore be designed so as to permit a large increase of temperature between inlet and outlet.

The Steam Temperature

12.184. The entry and exit temperatures of the coolant will determine, to a great extent, the best steam conditions to be used in the circumstances. For example, the temperature of the steam leaving the heat exchanger must be less than t_{out}, and the decision as to whether or not regenerative heating is to be used for the boiler-feed water will be based upon t_{in} (§ 12.189). The size and cost of the heat exchanger (boiler) required to produce a specified quantity of

steam will vary with both Δt_c and with the difference $t_{in} - t_{sat}$, where t_{sat} is the temperature of the saturated steam in the boiler, as shown qualitatively in Fig. 12.7. It is evident that the steam temperature will represent a compromise, since a high value of t_{sat}, which is desirable for high thermal efficiency, will mean an increase in the heat-transfer area (and cost) of the boiler, for a given t_{in}. Some advantage might be gained by adjusting the reactor conditions so as to increase t_{in}, but if t_{out} is fixed this would mean a decrease in Δt_c and, as a result, an increase in the boiler size (Fig. 12.7). In any event, a change in Δt_c would mean an adjustment of the flow rate of coolant in the reactor if the power level is to be maintained.

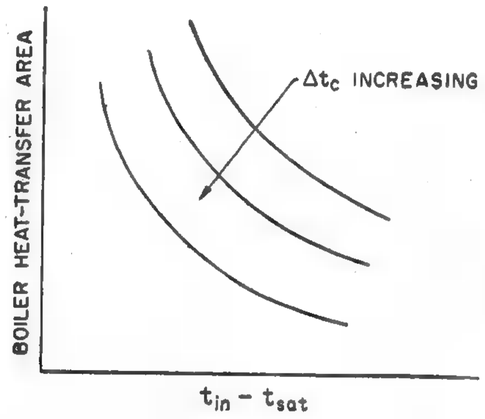


FIG. 12.7 Dependence of heat-transfer area in steam generator on system temperatures

12.185. It will be apparent from the foregoing remarks that there is an interdependence of the various temperatures in a reactor system, and the optimum conditions can be arrived at only by continuously repeating and refining the design calculations described above. The difficulty of the problem is accentuated by the fact that design changes which are beneficial from the heat-transfer standpoint are often found to be detrimental (and uneconomic) when the nuclear aspects are taken into consideration. Nevertheless, a satisfactory compromise, concerning coolant inlet and outlet temperatures and conditions in the boiler, will eventually be reached. As with engineering problems in general, increase in experience, which has hitherto been very limited, will facilitate arrival at a practical solution.

Homogeneous and Boiling Reactors

12.186. In homogeneous and boiling reactors the circumstances are somewhat different from those described above, which apply strictly to reactors with solid fuel elements. In a homogeneous reactor the question of a maximum rate of heat transfer from a solid surface does not arise and the operating temperatures are determined by the acceptable pressure in the reactor vessel and the conditions of the steam-turbine circuit. In a boiling-water reactor, which supplies steam directly to a turbine, the boiling temperature and the return temperature of the water are dependent on the steam-turbine system. However, the rate of heat transfer from the fuel elements to the boiling water, which depends upon the difference between the temperature of the fuel-element surface and the saturation temperature of the water (§ 11.183), must also be taken into consideration.

Temperatures with Water as Coolant

12.187. When water is the coolant the outlet temperature is determined by the acceptable pressure in the reactor vessel and the nature of the fuel-element cladding. If the latter is aluminum, the maximum permissible temperature of the water is probably about 400°F, but with zirconium (or zirconium alloy) an average outlet temperature of about 560°F could be tolerated. In order to prevent boiling of the water in the hottest cooling channel, the pressure in the reactor vessel would have to be about 2000 psi. The inlet (or return) temperature of the water to the reactor will be determined almost entirely by the characteristics of the steam-turbine system. A value 70° to 100°F below the reactor outlet is found, in general, to be satisfactory. The data in Table 12.4 are quoted as an example of the operating conditions of a proposed reactor using water under pressure as moderator and coolant.* The design is for an output of 246 Mw of electrical power, so that, with a heat-power of 1243 Mw, the expected over-all efficiency is about 20 per cent.

TABLE 12.4. PROPOSED OPERATING CONDITIONS FOR POWER REACTOR

Pressure vessel design, psi	~1000
Maximum fuel-element surface temperature, °F	500
Average coolant outlet temperature, °F	440.7
Coolant inlet (return) temperature, °F	369.4
Water velocity in reactor, ft/sec	25
Coolant flow rate, 10^6 lb/hr	54.75
Turbine throttle:	
Steam temperature °F	373.1
Steam pressure, psia	180
Reactor heat generation, Mw	1243

Temperatures with Sodium as Coolant

12.188. With sodium as coolant, the upper temperature limit is again dependent upon the fuel cladding; if this is stainless steel, the maximum permissible temperature of the liquid sodium is about 1200°F, and a reasonable maximum average temperature for the coolant leaving the reactor would appear to be around 1000° to 1050°F. This high temperature permits greater flexibility in the operation of the steam-turbine system, and upon this the temperature of the sodium returning to the reactor is largely dependent. A reasonable value for the difference between reactor inlet and outlet temperatures for the coolant would be about 300° to 400°F. With a coolant flow velocity of about 25 ft/sec in the reactor, the rate of heat removal would then be approximately the same

* Commonwealth Edison-Public Service Co. of Northern Illinois, Report CEPS-1111 (unpublished).

as for water, with a temperature change of about 100°F, in a reactor having the same flow area. The thermal efficiency would, of course, be appreciably greater with sodium, because of the higher steam temperature.

Steam Cycles for Nuclear Power

12.189. Since the conditions in a steam-turbine system have a bearing on the operating temperatures of the reactor, and vice versa, it is appropriate to refer briefly to the steam cycles that might be utilized for the production of electricity from the heat liberated in a nuclear reactor. In modern conventional central-power plants superheated steam is invariably used at the turbine throttle. There are, generally, up to four or perhaps five regenerative feed-water heaters and, possibly, one or two stages of reheat between high- and low-pressure turbines. The moisture content of the steam in the turbine must be kept low because an appreciable proportion of liquid water decreases the efficiency and causes increased erosion of the blades.

12.190. The adaptation of a steam cycle to the production of nuclear power is shown in the flow diagram in Fig. 12.8; only two regenerative feed-water heaters, although more are possible, and one stage of reheat are shown. In a water-cooled reactor, the temperature would probably not exceed 600°F, and, in these circumstances, superheating is difficult. It is possible that the use

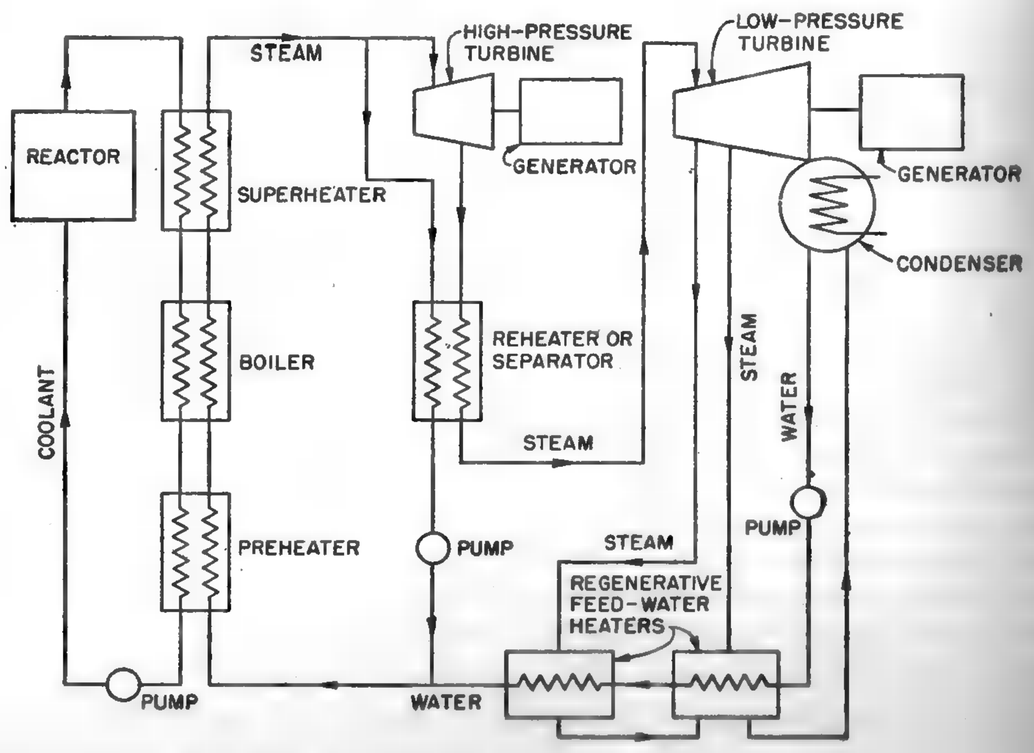


Fig. 12.8 Hypothetical steam cycle for nuclear power

of a moisture separator, instead of reheat, between the high- and low-pressure turbines might be advantageous in giving improved thermal efficiency at small cost. Regenerative feed-water heating is probably to be recommended, although the optimum number of stages used would have to be related to the reactor coolant inlet temperatures.*

12.191. When sodium is the coolant, with the possibility of obtaining steam at high temperatures, the steam-cycle conditions will resemble those of a conventional power plant to a considerable extent. The highest thermal efficiencies will probably then be obtained by using superheat, reheat (or moisture separation), and four or five stages of regenerative feed-water heating.

12.192. Since, at sufficiently high steam pressures, superheating is accompanied by a marked gain in efficiency, it has been suggested that a combustion-fired heater be used to superheat the saturated steam produced in the boiler associated with a water-cooled reactor. Whether this scheme would prove economically effective or not is uncertain because of the possible high cost of the superheater and of the combustible fuel.†

PRELIMINARY DESIGN CALCULATIONS

Fuel Loading and Core Dimensions

12.193. The final design of a nuclear reactor is attained only after numerous calculations and experimental observations. The latter may include not only criticality studies, as outlined in § 12.108, but also hydraulic measurements with a mock-up of the core, using inactive material, to study the flow pattern of the coolant and pressure drops within various parts of the system. It is expected that as knowledge and experience of reactor design increase, however, the need for a mock-up will decrease. Before proceeding to the experimental studies of reactor design problems, many calculations of a preliminary character must be made, and some of these will be indicated below.

12.194. Suppose it is required to design a cylindrical reactor to produce 1000 Mw of heat, using 1.2 per cent enriched uranium as fuel and ordinary water as moderator-coolant. As already seen, for such a reactor an average specific power of 24 kw/kg of uranium may be feasible, using plate-type fuel elements. Hence, to produce 1000 Mw, i.e., 10^6 kw, the reactor core should contain $10^6/24$, i.e., 4.2×10^4 kg or 46 tons of metal.

12.195. The average thermal flux can now be estimated to see that it has a reasonable value in view of the difficulties arising from radiation damage

* C. H. Robbins, *Nucleonics*, 11, No. 1, 42 (1953).

† C. H. Robbins, *loc. cit.*; Report on Atomic Power Symposium, Chalk River, Ont., 1953 (CRR-548-A), Chapter 11 (by I. N. MacKay). According to a private communication from J. A. Lane, studies at Oak Ridge National Laboratory indicate that power costs in a plant using a combustion-fired superheater in conjunction with a reactor are not appreciably lower than for a nuclear power plant without the superheater. The combination may, nevertheless, be desirable for other reasons.

and xenon poisoning at high fluxes. Utilizing equation (12.11.1), with $P = 10^3$ Mw $= 10^9$ watts, $w = 500$ kg $= 5 \times 10^5$ grams, and $\sigma_f = 549$ barns for uranium-235, it follows that

$$\phi = \frac{(1.2 \times 10^{13})(10^9)}{(5 \times 10^5)(549)}$$
$$= 4.3 \times 10^{13} \text{ neutrons/(cm}^2\text{)(sec)}.$$

With this value of the thermal-neutron flux, the equilibrium value of the xenon poisoning during operation will be about 0.04 and the maximum after shutdown about 0.13.

12.196. The density of uranium metal is 19 g/cm^3, and hence the volume occupied by the 4.2×10^4 kg of fuel is 78 ft^3. In addition, the core will contain about one third of this volume, i.e., 26 ft^3, of cladding and structural material for the fuel elements. Approximate criticality calculations indicate that for the estimated mass of 46 tons of fuel to have an effective multiplication sufficiently larger than unity to allow for poisoning, temperature increase, etc., the volume of the cylindrical core will be roughly 320 ft^3. The volume of the water is then equal to this volume minus the volume of the fuel elements (i.e., $78 + 26 = 104$ ft^3) and the volume required for insertion of the control rods. The water volume is thus about two thirds of the total volume of the core. The power (heat) density of the reactor will be $10^6/320$ kw/ft^3, i.e., 1.07×10^7 Btu/(hr)(ft^3) or 110 kw/liter (cf. Table 11.1).

12.197. A cylinder 7.00 ft in diameter and 8.32 ft in height will have a volume of 320 ft^3 and so these may be taken as reasonable dimensions for the reactor core. The circular cross-sectional area is thus 38.4 ft^2 and the total flow area of the water will be roughly two thirds of this, namely, 25 ft^2. The total cross-sectional area of the uranium fuel would be (38.4)(78)/(320), i.e., 9.35 ft^2. The 3 ft^2 or so occupied by the cladding-structural material may be neglected as being "dead space," provided that neutron capture by the cladding on the fuel is quite small, as it usually will be.

Number and Spacing of Fuel Plates

12.198. In order to sustain the postulated specific power, the fuel region of the plate should be no more than 0.2 in. thick. Upon performing the subsequent calculations on the basis of this thickness it is found that the fuel-surface temperature is higher than is considered desirable, so that thinner fuel elements are necessary. Suppose the thickness chosen is 0.15 in.; if each plate is 3 in. wide, the total number of fuel plates is given by

$$\text{Number of fuel plates} = \frac{(9.35)(12)^2}{(0.15)(3.0)} = 3000.$$

12.199. Because the ratio of the volume of water to that of fuel in the reactor is 25 to 9.35, it follows that the distance between the fuel plates of thickness

0.15 in. must be

$$\text{Distance between fuel plates} = \frac{(0.15)(25)}{9.35} = 0.40 \text{ in.}$$

The equivalent diameter D_e of the coolant passage, which is twice the distance between the plates, is consequently 0.80 in.

12.200. Since each plate is 8.32 ft long and 3 in. wide and has two exposed faces, the heat-transfer area per plate, i.e., A_h, is (2)(8.32)(3/12) = 4.15 ft^2. The average heat-release per plate is $(10^6)(3.413 \times 10^3)/3000 = 1.14 \times 10^6$ Btu/hr; and so the average heat flux in the reactor is

$$\text{Average heat flux} = \frac{1.14 \times 10^6}{4.15} = 274{,}000 \text{ Btu/(hr)(ft}^2\text{)}.$$

The maximum heat flux may be three or four times this amount, but it will probably not exceed the generally accepted limiting practical value of 10^6 Btu/(hr)(ft^2) at which burn-out may occur.

Calculation of Heat-transfer Coefficient

12.201. With these data available, it is now possible to proceed with the direct heat-transfer calculations. Suppose the average rise in temperature, Δt_c, of the coolant in passing through the reactor is to be 75°F (§ 12.187), the inlet temperature being 375°F and the outlet temperature 450°F. The mass flow rate, w, of the water can then be determined from equation (11.137.1) in the form

$$w = \frac{q}{c_p \Delta t_c},$$

where q is 10^6 kw $= 3.413 \times 10^9$ Btu/hr and Δt_c is 75°F. The specific heat of water at the mean temperature of 410°F is 1.06 Btu/(lb)(°F), so that

$$w = \frac{3.413 \times 10^9}{(1.06)(75)} = 42.9 \times 10^6 \text{ lb/hr}.$$

12.202. The mass flow rate is related to the coolant velocity, v, by the relationship $w = v\rho A_f$ or

$$v = \frac{w}{\rho A_f},$$

where A_f is the appropriate flow area for the coolant. It was calculated above that the total flow area of the water is 25 ft^2, and this would be the value of A_f if a single pass of coolant is employed. It was seen in § 11.45, however, that it may be advantageous to design the flow system so that two or more passes occur. Hence, it will be postulated that there are two passes, the coolant flowing up the center of the reactor core and down the outer regions, with half the total flow area being involved in each pass. The flow area, A_f, per pass is thus 12.5 ft^2 and, hence, since the density of water at 410°F is 53.6 lb/ft^3, the coolant

flow velocity in each pass is

$$v = \frac{42.9 \times 10^6}{(53.6)(12.5)} \text{ ft/hr}$$
$$= 17.9 \text{ ft/sec.}$$

This flow velocity is somewhat less than the acceptable maximum value of 25 to 35 ft/sec and it might perhaps be increased with advantage, e.g., by decreasing the flow area in the core or the temperature drop, Δt_c. There would be an increase in the heat-transfer coefficient, although it would be accompanied by an increase in the pumping power required for the coolant.

12.203. Upon inserting the value for v (in ft/sec) and that for D_e (in inches), namely, 0.80 in., obtained above, into equation (11.128.1), the average heat-transfer coefficient (at 410°F) under the given conditions is found to be

$$h = 170[1 + (10^{-2})(410) - (10^{-5})(410)^2] \frac{(17.9)^{0.8}}{(0.80)^{0.2}}$$
$$= 6100 \text{ Btu/(hr)(ft}^2\text{)(°F)},$$

which would appear to be satisfactory for the present purpose.

Maximum Temperature of Fuel-Element Surface

12.204. Since in its first pass the coolant will flow through the center of the reactor where the flux is highest, it may be supposed that about two thirds of the total heat generated in the reactor will be removed in this pass.* Hence, for the first pass q may be taken to be 660 Mw or $(660)(3.413 \times 10^6) = 2.25 \times 10^9$ Btu/hr; since half the fuel plates, i.e., 1500 plates, are involved, the value of q per plate is 1.50×10^6 Btu/hr. The heat transfer area, A_h, per plate is 4.15 ft^2, and, hence, Δt_s, the average temperature drop between the surface of the fuel plate and the coolant, is given by equation (11.67.1) as

$$\Delta t_s = \frac{q}{hA_h}$$
$$= \frac{1.50 \times 10^6}{(6100)(4.15)} = 60°\text{F}.$$

The maximum value of this temperature drop in the central region of the reactor, excluding the effects considered in the next paragraph, is probably 1.6 or so times as large as the average value, so that $(\Delta t_s)_{max}$ is nearly 100°F.†

12.205. The foregoing calculations are based on the tacit assumption of strict regularity of the coolant flow channels and of uniformity of flow distribution. In actual practice deviations are bound to occur and in order to allow for them

* The exact fraction can be calculated from the known power distribution, but the value postulated here is sufficiently accurate for the present purpose.

† A more precise method of determining $(\Delta t_s)_{max}$ would be to use the procedure described in § 11.102, *et seq.*; this would require a knowledge of the power distribution in the reactor. For preliminary purposes, however, the calculation given above is adequate.

the so-called "hot channel" factors have been introduced. These safety factors are entirely empirical in nature and vary from one reactor to another. The data in Table 12.5,* which have been proposed for a water-cooled reactor with highly enriched fuel, are quoted in order to give some indication of the accepted magnitudes of hot-channel factors due to various causes. Thus, in estimating the maximum value of Δt_s for a hot channel, the result calculated above should be multiplied by the applicable factors in the first column of the table. In the present case, the enrichment variation is probably not significant and so the over-all hot-channel factor is about 1.6; the maximum hot-channel value for Δt_s, thus (1.6)(100) = 160°F.

TABLE 12.5. HOT-CHANNEL FACTORS

Effect	Δt_s	Δt_c	Q
Inability to predict flux	1.20	1.20	1.20
Variation in enrichment	1.20	1.10	1.20
Variation in channel diameter	1.25	1.40	1.10
Nonuniform flow distribution	1.02	1.02	—
Warping of fuel element	1.03	1.04	—

12.206. The total increase in the temperatures of the coolant in the first pass is expected to be two thirds of 75°F, i.e., 50°F. The average increase is then about 25°F; upon multiplying this by 1.6, to obtain the normal maximum, and by the appropriate hot-channel factor from the second column of Table 12.5, the maximum hot-channel value of Δt_c is estimated to be about 65°F. Consequently the maximum hot-channel temperature at the fuel-element surface is expected to be

$$375 + 160 + 65 = 600°\text{F},$$

where 375°F is the temperature at which the coolant enters the reactor. This is sufficiently less than the maximum acceptable temperature for zirconium (§ 12.168) to make the latter a satisfactory cladding material. To prevent the water from boiling at the fuel-element surface, the system would have to be pressurized to at least 1600 psia.

Maximum Temperature within Fuel Element

12.207. Disregarding the temperature drop through the cladding, which will be quite small, the average temperature drop across the fuel can be obtained from equation (11.82.1), i.e.,

$$t_0 - t_1 = \frac{Qa^2}{2k},$$

where a is the half-thickness of the fuel. For the central region of the reactor, in which the first pass of the coolant occurs, q is 2.25×10^9 Btu/hr and since

* Unpublished information from Argonne National Laboratory.

the volume of the fuel is $(\frac{1}{2})(78) = 39$ ft³, the average value of Q is 5.8×10^7 Btu/(hr)(ft³). This must be multiplied by factors allowing for the theoretical ratio of maximum-to-average heat-source strength and for the hot-channel effect, as given in the third column of Table 12.5. The appropriate value of Q for calculation purposes is thus about 1.2×10^8 Btu/(hr)(ft³). Since k for uranium is 14 Btu/(hr)(ft²)(°F/ft) and a is 0.075 in., it follows that

$$(t_0 - t_1)_{max} = \frac{(1.2 \times 10^8)(0.075/12)^2}{(2)(14)}$$
$$= 170°F.$$

The maximum (hot-channel) temperature within the uranium fuel element is thus 600 + 170 = 770°F, which is considerably below the safe limit (§ 12.115).

12.208. A similar study for the second pass shows that the maximum hot-channel temperatures do not exceed those derived above for the central portion of the reactor. It may be concluded, therefore, that the proposed reactor design will form a satisfactory basis for further study. A more detailed correlation of the reactor system with the steam-turbine system and the results obtained from exponential, critical, and hydraulic experiments with a core mock-up will permit development of the details of the final design.

12.209. It should be mentioned that the procedure just described, which commenced with an assumed reasonable value of the specific power (§ 12.194), is not the only method for approaching the reactor design problem. In some cases, the design may begin with dimensional restrictions; for example, if the reactor is to be mobile or transportable. In others, the starting point may be the maximum surface temperature. However, in all instances, the same general treatment is involved, although the calculations are not necessarily performed in the order given above.

Changes in Thermal Design Parameters

12.210. In the foregoing discussion the only design parameter changed in the course of the calculations was the thickness of the fuel plates, from 0.20 to 0.15 in., because the preliminary estimates showed that the larger value would lead to unacceptable temperatures. In general, a decrease in the thickness of the fuel element, since it is accompanied by an increase in the heat-transfer area, results in a decrease in the temperature difference between the metal surface and the coolant and in the temperature drop within the fuel element itself. Both of these changes are advantageous in providing increased flexibility in other aspects of the design problem. There are, however, other ways in which the reactor design may be changed to provide more desirable heat-transfer conditions.

12.211. An increase in the mass flow rate of the coolant, for example, without change in the heat-transfer area or flow cross section, will result in an increase in the heat-transfer coefficient due to the increase in the flow velocity. The same result can be achieved by keeping the mass flow rate and heat-transfer area constant and decreasing the flow area. This is essentially what happens when the number of passes of the coolant through the reactor is increased by the use of baffles. These changes would be more beneficial for water as coolant than for a liquid metal, since for the latter the effect of flow velocity on the heat-transfer coefficient is relatively small. It should be noted that all the design changes considered above are accompanied by an increase in the pressure drop of the coolant and, hence, by an increase in the pumping power required. This drawback may or may not be significant in comparison with the advantages to be gained.

12.212. The maximum power output of the reactor can be increased by decreasing the ratio of maximum to average heat flux by making the volumetric heat-production rate more uniform throughout the core, i.e., by "flattening" the neutron flux, as a result of an appropriate distribution of the fuel or by the use of neutron absorbers (see § 11.44). The temperature rise of the coolant is then almost the same in all the channels and higher specific powers can be attained. Uniform temperature rise can also be achieved by appropriate orificing of the coolant channels, i.e., by adjusting the rates of flow of the coolant through the channels, so that the temperature rise is almost the same in each one (§ 11.45).

ECONOMIC ASPECTS OF NUCLEAR POWER *

Review of Cost Estimates

12.213. A number of estimates have been published of the capital costs of large-scale (electrical capacity of 100 Mw or more) nuclear power plants and of the cost of electricity at the bus bar of these plants, but they vary over a wide range.† There are several reasons for the considerable variation in the conclusions reached. In some cases the estimates are high because some research and development costs have been included and because conservative assumptions were made with regard to technical and nontechnical economic factors. On the other hand, the lower estimates are postulated upon the realization of all foreseeable advances in technology, and so are applicable to nuclear power plants

* This section is based on a review prepared by J. A. Lane (private communication).

† W. H. Zinn, *Nucleonics*, **10**, No. 9, 8 (1952); C. Starr, *ibid.*, **11**, No. 1, 62 (1953); J. A. Lane, *ibid.*, **12**, No. 6, 12 (1954); **13**, No. 1, 24 (1955); J. A. Lane and S. McLain, *Chem. Eng. Prog.*, **49**, 287 (1953); W. E. Abbott, *ibid.*, **50**, 245 (1954); D. E. Ferguson, *Scientific Monthly*, **79**, 125 (1954); F. K. McCune, Atomic Industrial Forum Report, July 1954, p. 62; T. Stern, *Chem. Eng. Prog. Symposium Series*, No. 11, **50**, 177 (1954); T. T. Shimazaki, *ibid.*, No. 12, **50**, 113 (1954); C. A. Rennie, *ibid.*, p. 222; see also, "Reports to the U. S. Atomic Energy Commission on Nuclear Reactor Technology," U. S. Government Printing Office, May, 1953; *Nucleonics*, **11**, No. 6, 49–64 (1953); Commonwealth Edison-Public Service Co. of Northern Illinois, Report CEPS-1111 (unpublished); Foster Wheeler-Diamond Alkali-Pioneer Service Nuclear Power Project, Report NEA-5301 (unclassified summary); "Atomic Energy in Industry," 3rd Annual Conf., National Industrial Conference Board Proceedings, 1955.

which might be built after more experience has been gained in their design, construction, and operation. However, if all the data are reduced to a comparable basis, as they are in Fig. 12.9, the general conclusion to be drawn is that, after the advantages of expected improvements in technology have been realized, the cost of nuclear power is estimated to be 6 to 7 mills/kw-hr for all five types of reactors which appear to show promise for the near future.

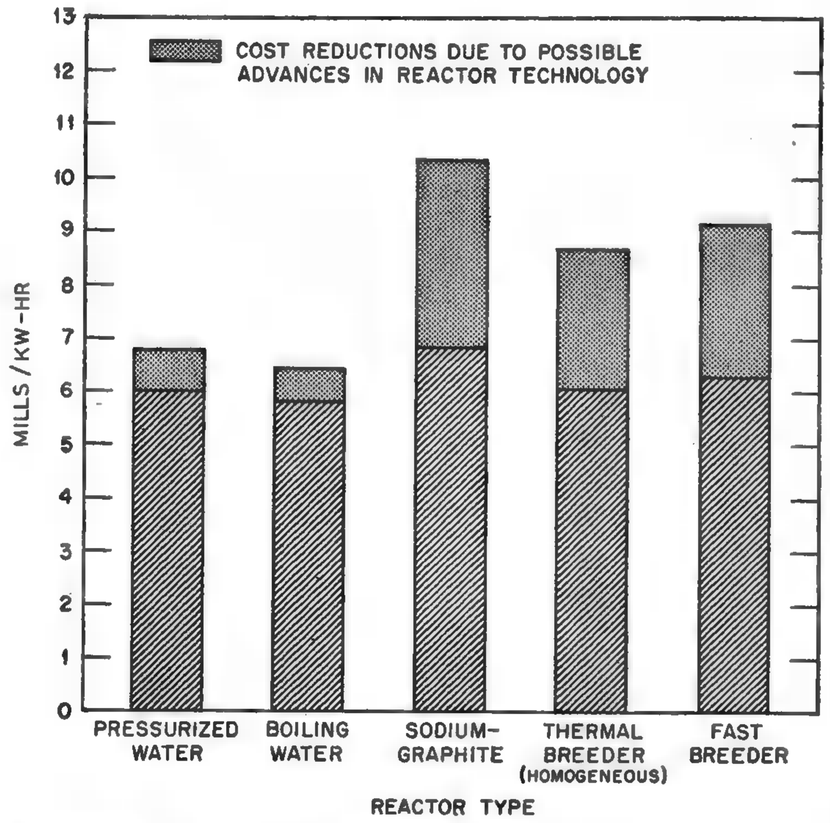


FIG. 12.9 Comparison of cost of electricity from reactors of various types. (J. A. Lane)

12.214. In all cases, however, the conclusions are greatly influenced by such factors as safety precautions, reactor lifetime, fuel-element lifetime, continuity of plant operation, and the charges to be made for nuclear fuels and other special reactor materials. It would appear to be impossible, therefore, to make a realistic estimate of costs without actual reactor construction and operating experience, and without more definite information concerning the charges for special materials which are under government control. This means that it is not possible to decide from estimates based upon assumed capital costs, operating conditions, and accounting procedures whether or not nuclear power will be competitive with power from conventional (fossil) fuels. Such estimates do, however, provide the groundwork for establishing the conditions under which nuclear-fueled plants will be competitive with those using coal or oil as fuel.

Comparison of Nuclear and Conventional Power Plants

12.215. There are a number of special features of a nuclear power plant which influence the cost of power produced, and these will be reviewed briefly. First, the radioactivity in the nuclear reactor plant represents a hazard not encountered in conventional power plants. As seen in Chapter IX, all necessary precautions must be taken to reduce this hazard to the point where it can be tolerated by operating personnel and where it will not represent any inconvenience or danger to the surrounding population. This means added construction costs for shielding and to prevent the escape of radioactive materials; there are also increased operating costs because of the necessity for regular inspection and because of the difficulties associated with handling materials and maintaining equipment.

12.216. Second, the capital cost of a nuclear plant includes that of special materials, e.g., heavy water, zirconium, high-purity graphite, sodium, and (possibly) titanium. The total capital investment in a nuclear plant may thus be higher than in a coal-fired plant. However, this increase in cost may be partially offset by the decrease due to the reactor and heat exchanger (steam generator) being smaller in size than the corresponding boiler and coal-handling facilities (see Fig. 1.1).

12.217. Third, in general, nuclear plants will produce steam at somewhat lower temperatures and pressures than modern plants using fossil fuels, and so the former will have lower thermal efficiencies. The possibility of reducing the net cost of nuclear fuel, by conversion or breeding, to the point where it is considerably less than that of conventional fuel, however, makes the difference in thermal efficiencies less significant. The net effect of lower steam temperatures in nuclear plants on the cost of power may thus not be great.

12.218. Finally, the sale of fissionable material and of isotopes produced in a nuclear reactor may have an important bearing on nuclear power economics. In this connection, the development of uses for mixed or isolated fission products, which appears probable (§ 7.215, *et seq.*), may provide an additional source of revenue.

12.219. Because of these differences between nuclear and conventional power plants the distribution of capital charges and operating costs will not be the same, even though the ultimate power costs are comparable. In Table 12.6 there is given what appears to be a reasonable breakdown of the costs of a (heavy-water moderated) nuclear plant with an electrical capacity of 200 Mw, based on the *assumption* that power can be produced at the bus bar for 6.7 mills/kw-hr. The capital cost, apart from fuel and special materials, is seen to be $190/kw. A conventional plant, producing power for the same price, should cost $150/kw.

Factors Influencing Nuclear Power Costs

12.220. It should be emphasized that the costs quoted in Table 12.6 for a large-scale nuclear plant are based on the somewhat arbitrary choice of 6.7 mills/kw-hr, which is the average cost of power from coal-fired plants. In actual cases the costs will undoubtedly differ from these values, depending upon

TABLE 12.6. ESTIMATED COSTS OF PLANTS OF 200 MW CAPACITY BASED ON ASSUMED 6.7 MILLS/KW-HR FOR ELECTRICITY

	Nuclear Power Plant		Conventional Plant	
	Capital* Cost ($/kw)	Power Cost (mills/kw-hr)	Capital* Cost ($/kw)	Power Cost (mills/kw-hr)
Capital Items				
Reactor structure and controls	25			
Reactor building	10			
Heat-removal equipment	35			
Turbine plant	70			
Turbine plant building and site facilities	30			
Miscellaneous electrical equipment	20			
Total	190	4.1	150	3.2
Inventory Items				
Nuclear fuel	30			
Special materials (heavy water)	30			
Total	60	1.1		
Operating Items				
Fuel consumption		0.5		2.8
Operation and maintenance		1.0		0.7
Total power cost at bus bar		6.7		6.7

* Based on 80 per cent load factor and 15 per cent annual charges.

the type and design of the reactor, and on the total power output of the plant, as well as on various technical and nontechnical factors, such as safety, continuity of operation, and material costs. The influence of these factors on nuclear power costs is discussed below.

Reactor Type

12.221. It is apparent from Fig. 12.9 that, for the systems which now appear to be the most promising, and assuming that reasonable advances in technology

will be realized, the cost of power produced will not be greatly dependent upon the type of reactor. Although some variations in cost are indicated, it is not possible to say, until further experience has been gained, whether they are significant or not. In making these estimates it is assumed, of course, that the particular designs represent the optimum combination of nuclear and heat-transfer requirements. It should be mentioned, finally, that new technical discoveries, at present unforeseen, may change the situation with regard to nuclear power costs, although it does not seem probable that these changes will be very great.

Power Plant Capacity

12.222. Because of the decrease in capital charges per kilowatt-hour, it is likely that the cost of electricity will decrease with increasing power output of the plant. In very large nuclear plants, the cost of power may be small, but such plants must be regarded as competing with conventional plants of large base-load which already produce power at low cost, e.g., 3 to 4 mills/kw-hr. At the other extreme, small nuclear plants, located in remote areas, may produce power for 15 to 20 mills/kw-hr and yet still compete with small diesel plants or coal-steam plants. The reason for this is the high cost of transporting fuel for conventional power plants as compared with the negligible cost for nuclear fuel.

Safety Precautions

12.223. According to the formula given in § 9.7, a nuclear power plant would require about an acre of exclusion area for each kw of installed electrical capacity. This might add from 2 to 8 mills/kw-hr to the cost of power, depending upon the value of the land, and so would make nuclear power uneconomical. An alternative approach is to put the reactor underground, as is being done with the PWR at Shippingport, Pennsylvania, or provide a building which would contain any radioactivity that might escape from the reactor in the event of an accident. It has been estimated that this type of precautionary measure will add something of the order of $1,000,000 to the cost, but for a large-scale plant this would represent no more than a per cent or so of the total capital outlay.

Continuity of Operation

12.224. The reliability with which the reactor operates, with respect to supplying the load demand, and the lifetime of the reactor and its associated equipment are important economic considerations. Since experience in the operation of nuclear power plants is lacking, it has been assumed in various cost studies that the reactor will last anywhere from 5 to 20 years and operate at load factors ranging from 70 to 90 per cent. Since capital costs of nuclear plants are likely to be high, it is necessary to achieve long reactor life and high load factors if power is to be produced economically. At the present time it is not possible to say to what extent these aims will be achieved.

Cost of Special Materials

12.225. Essentially all power reactors, of no matter what design, will require some special material or another. The cost of zirconium (or possibly titanium) as fuel cladding, heavy water, beryllium, or high-purity graphite as moderator, and sodium as coolant, will have an important influence on nuclear power economics. However, in well-designed reactors of high power density the cost per kilowatt-hour of such materials can probably be kept within reasonable limits. In any event, with increasing production experience and demand for these substances, the cost will probably decrease in time.

Cost of Nuclear Fuel

12.226. There are two distinct aspects of the cost of nuclear fuel which must be considered: first, the capital cost of the fuel inventory and the corresponding annual charges and, second, the cost of the fuel (fissionable material) actually consumed in the reactor. Since all fissionable and fertile materials are under government control, it may be presumed that the fuel will be loaned to the power plant which will pay a rental charge for its use. If this charge is kept as low as 4 or 5 per cent, fissionable and fertile material inventory costs should be quite moderate in properly designed systems. The inventory cost given in Table 12.6 is based on an assumed charge of 10 per cent on the capital value of the fuel.

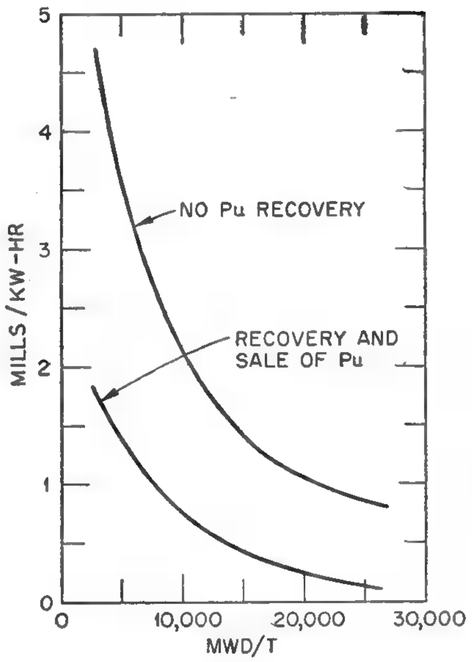


FIG. 12.10 Effect of burn-up on cost of nuclear fuel per kilowatt-hour of electricity. (J. A. Lane)

12.227. The net cost of the fuel consumed in a nuclear plant is a function of many interrelated variables, e.g., cost of ore, extraction costs, type of fuel, degree of enrichment, fuel-element fabrication costs, fuel-cycle time, i.e., degree of burn-up, cost of processing spent fuel elements and fertile material from breeder, and, finally, thermal efficiency of the power system.* In any given case it is necessary to consider all these factors and to balance the fuel costs against fixed costs, as is done in conventional power plants.

12.228. Some indication of the effect of burn-up on the cost of fuel consumed in a power reactor is given by the curves in Fig. 12.10; the fuel is slightly enriched uranium (1 to 2 per cent uranium-235), this being an attractive material for

* Cf. W. H. Zinn, *Nucleonics*, **10**, No. 9, 8 (1952); M. Benedict, *Ind. Eng. Chem.*, **45**, 2372 (1953); T. M. Snyder, *Proc. Conf. on Nuc. Eng.*, Berkeley, 1953, C-11.

various types of reactors. The basis of the calculations is a cost of \$15/lb for U_3O_8 with credit for unconsumed uranium-235, and a thermal efficiency of 25 per cent. The upper curve in Fig. 12.10 refers to the case in which there is no recovery of the plutonium-239, produced by conversion of the uranium-238, and the lower for a system in which a credit of \$20/gram, which is essentially its fuel value (§ 12.89), is applied for the plutonium-239 recovered. The burn-up of fuel is expressed in terms of megawatt-days of exposure in the reactor per ton of fuel, represented by MWD/T.

12.229. The cost of the nuclear fuel consumed as given in Table 12.6, on the basis of an assumed cost of 6.7 mills/kw-hr for power, is only 0.5 mill. This means that, for the conditions to which Fig. 12.10 applies, a fuel burn-up equivalent to about 14,000 MWD/T must be attained, assuming recovery and sale of the plutonium produced. If the reactor contains 50 tons of fuel, which is not unreasonable, this exposure corresponds to a fuel cycle of nearly two years, i.e., the reactor would have to be reloaded every two years, assuming a (heat) power output of 1000 Mw.

12.230. In a breeder reactor, in which there is no net consumption of fuel, costs are dependent upon the expense of recovering the fuel from the spent elements and the fissionable species from the fertile blanket. It has been estimated that processing costs of enriched material must be reduced to the order of \$1 to \$2/gram of fuel if economic operation is to be achieved.

Cost of Reactor Components and Associated Equipment

12.231. Because of the nature of most nuclear systems, the reactor components and other equipment have more in common with those encountered in a chemical plant than in a conventional power station. This is because the primary heat-transfer medium is either a high-pressure coolant, a liquid metal, or a somewhat corrosive solution or slurry. The handling of such materials involves, in all cases, the use of stainless steel or more expensive metals, and special equipment, such as canned-rotor and electromagnetic pumps, leak-proof heat exchangers, special valves, and large pressure vessels. To reduce the cost of this equipment will require the development of an industry to produce the required items on a more-or-less routine basis. Even if this possibility is realized, the achievement of economic nuclear power will depend ultimately upon the ability of the engineer to design and construct an efficient and reliable reactor system.

SYMBOLS USED IN CHAPTER XII

A_f	coolant flow area
A_h	heat-transfer area
a	radius of cylindrical fuel element
a	half-thickness of fuel plate
B^2	buckling
C	conversion ratio

c_p	specific heat
D_e	equivalent diameter of coolant passage
g	weight of fissionable material in grams
H	effective (over-all) heat-transfer coefficient
h	heat-transfer coefficient
k	thermal conductivity
P	reactor power
p	resonance escape probability
Q	volumetric heat source (rate of heat removal or production per unit volume)
q	rate of heat removal from reactor
q_{max}	maximum rate of heat removal
r	isotope ratio (N_{235}/N_{238})
t_0	temperature in interior of fuel element
t_f	temperature of coolant fluid
t_{in}	inlet temperature of coolant to reactor
t_{lim}	limiting temperature in reactor
t_{out}	outlet temperature of coolant from reactor
t_{sat}	saturation temperature of water
Δt_c	increase in temperature of coolant
Δt_s	temperature drop from fuel element surface to coolant
$(\Delta t_s)_{max}$	maximum surface temperature drop
v	flow rate of coolant
w	mass flow rate of coolant
α	ratio of capture (nonfission) to fission cross sections
ϵ	fast-fission factor
η	fission neutrons liberated for each neutron absorbed in fuel
ν	fission neutrons liberated for each neutron causing fission
ρ	density
Σ_{235}	macroscopic absorption cross section of uranium-235
Σ_{238}	macroscopic absorption cross section of uranium-238
σ_{235}	microscopic absorption cross section of uranium-235
σ_{238}	microscopic absorption cross section of uranium-238
σ_f	microscopic fission cross section
τ	Fermi age of (resonance) neutrons
ϕ	neutron flux

PROBLEMS

1. Compare the heat-transfer and nuclear properties of a natural-uranium fuel element in the form of a cylinder 0.5 in. in diameter clad with a 0.02-in. thickness of aluminum, with a 4-in. wide plate-type element of the same cross-sectional area and having the same thickness of cladding.

2. A reactor, employing natural uranium as fuel and heavy water as moderator and coolant, is to have a heat power output of 200,000 kw. The cylindrical fuel rods are to have a diameter of 0.25 in., with an aluminum cladding 0.02 in. thick, and the heavy water is to enter the reactor at 400°F and to leave at 500°F. Assuming that the weight ratio of heavy water to fuel in the reactor, needed to satisfy nuclear requirements, is 2 to 1, perform the preliminary design calculations. Comment on the effectiveness of this reactor and indicate directions in which improvements might be possible.

3. Estimate the value of the initial conversion ratio for the reactor described in the preceding problem.

4. A sodium-cooled, graphite-moderated reactor is to use stainless-steel clad, plate-type fuel elements of 2.0 per cent enriched uranium. The liquid sodium is to enter the reactor at 700°F and leave at 1000°F, and the heat power output is to be 100 Mw. Assuming that the weight of graphite must be three times that of the fuel, develop a preliminary design for this reactor and its associated turbogenerator steam system.

5. An alternative to a two-pass flow system for the coolant would be to use a single pass but to vary the composition of the fuel in different parts of the core. Suggest a reactor design based on this concept and consider its advantages and disadvantages.

6. The following suggestions have been put forward as possible bases for reactor designs: (*a*) heavy water moderator and liquid sodium as coolant and (*b*) steam at high temperature as the (gaseous) coolant. Develop possible designs and operating conditions and comment on the value of the suggestions.

Chapter XIII *

DESCRIPTIONS OF NUCLEAR REACTORS

Introduction

13.1. The short descriptions given below are restricted to reactors which already exist or will definitely be completed in the near future. Reactors that are still in the conceptual design stage, such as those proposed by the various industrial groups (§ 1.23) and the Brookhaven LMFR (§ 12.141), will not be described here. Although some of the material contained in this chapter has been given in earlier sections of the book, it seemed worth while to collect the available information in one place for convenience.

13.2. Most of the reactors described have been designed for experimental purposes, being either research reactors or reactor experiments, and few are of advanced design. Although the EBR-1 and the HRE-1 have generated electrical power the quantities are not large. The only reactors definitely known to be capable of producing substantial amounts of power by early 1955 are the land-based prototype of the STR and its successor installed in the *U.S.S. Nautilus*, but little information concerning the design of these reactors has been released. In spite of the limitations, it is hoped that the descriptions and figures which follow will give some indication of how certain design considerations have been implemented in reactor construction.†

13.3. The nature of the moderator will be taken as a basis for classification; thus, reactors moderated by (1) graphite, (2) heavy water, and (3) ordinary water will be considered in this order. This will be followed by a reference to unmoderated, i.e., fast, reactors. To a large extent the order adopted follows the historical development of nuclear reactor design.

Graphite-moderated Reactors

Chicago Pile-1 (CP-1) ‡

Fuel: about 40 tons natural uranium (6 tons as metal, the remainder as oxide, mainly UO_2 and some U_3O_8)

* Reviewed by J. A. Lane and N. F. Lansing.

† For tabulated summaries of the important characteristics of many reactors, see H. S. Isbin, *Nucleonics*, **10**, No. 3, 10 (1952); **11**, No. 6, 65 (1953).

‡ E. Fermi, Report AECD-3269; *Amer. J. Phys.*, **20**, 537 (1952); see also Report TID-202.

Moderator: 385 tons graphite (including 92 tons reflector)
Reflector: 1 ft graphite
Shield: none
Over-all dimensions: $24\frac{1}{2} \times 24\frac{1}{2} \times 19$ ft high
Controls: 1 safety, 1 shim, 1 regulating (cadmium strip or boron steel)
Coolant: ambient air
Power: 0.5 watt (operated at 200 watts for short time)

13.4. The first self-sustaining nuclear chain reaction was realized in the Chicago Pile-1 (CP-1) on December 2, 1942; this natural uranium-graphite reactor was constructed on a squash court under the West Stands of Stagg Field, University of Chicago. The structure consisted of crisscross layers of

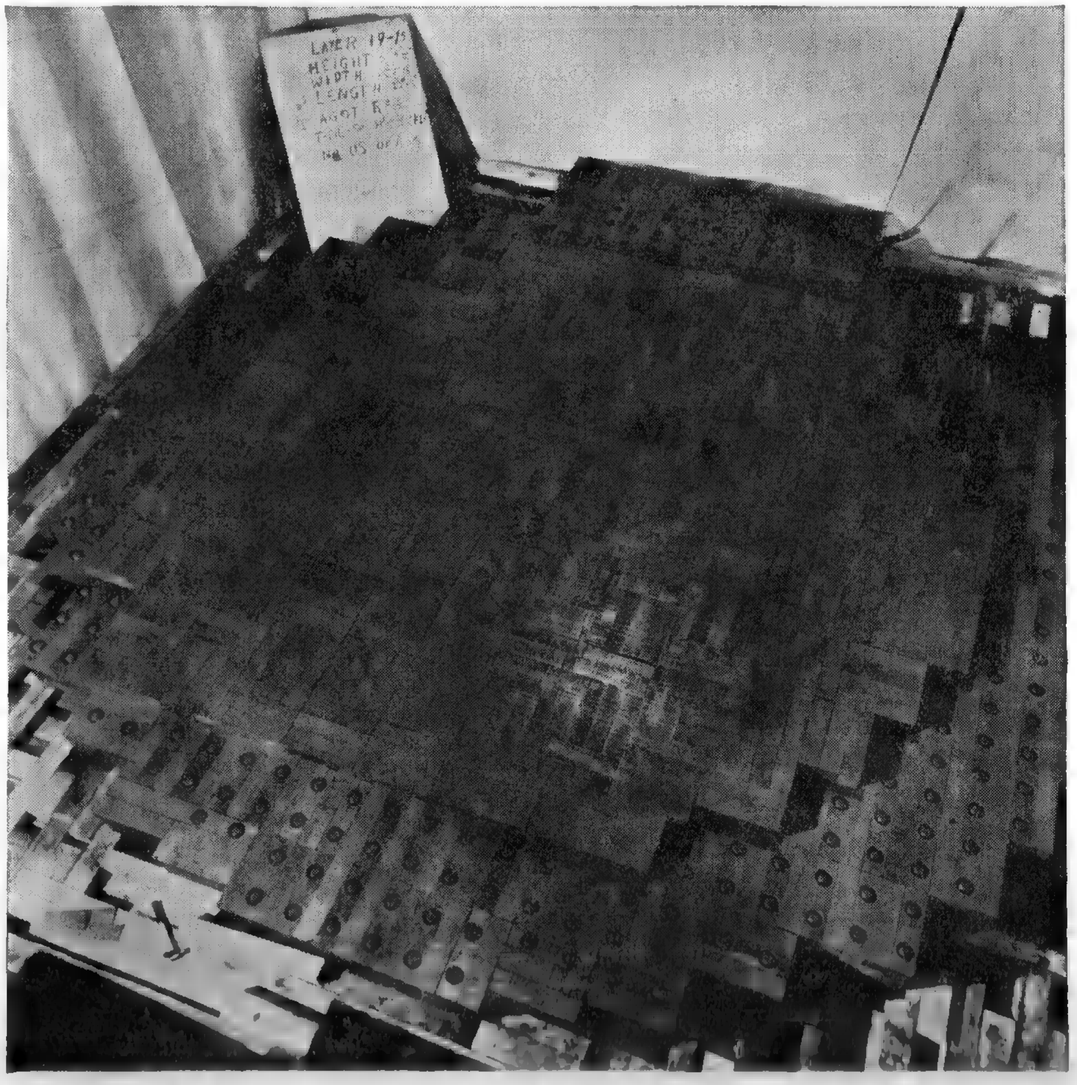


FIG. 13.1 CP-1 during construction, showing 18th layer containing fuel lumps and 19th layer of graphite only. (Argonne National Laboratory)

graphite blocks, approximately $4\frac{1}{8} \times 4\frac{1}{8} \times 16\frac{1}{2}$ in. in size; holes were made $8\frac{1}{4}$ in. apart in some of the blocks in alternate layers, and in these were placed a total of nearly 20,000 short cylinders of either uranium metal ($2\frac{1}{4}$ in. diameter by $2\frac{1}{4}$ in. long) or oxide (3 in. × 3 in.). The size of the fuel lumps and their spacing, i.e., a square lattice with an $8\frac{1}{4}$-in. pitch, were chosen to give the maximum value of the infinite multiplication factor (§ 3.177) in order to minimize the uranium requirements. The eighteenth layer, containing fuel, and the nineteenth layer, consisting of graphite alone, are seen in Fig. 13.1, which is the only photograph taken during the construction of CP-1. It was because the layers were piled one upon the other that the name "pile" was adopted to describe the arrangement.

13.5. According to the original design calculations, the active part of the reactor was to have been spherical, but it became critical sooner than expected. Consequently the final shape approached that of a sphere with a flattened top. The control rods were located in horizontal slots in the graphite layers and were

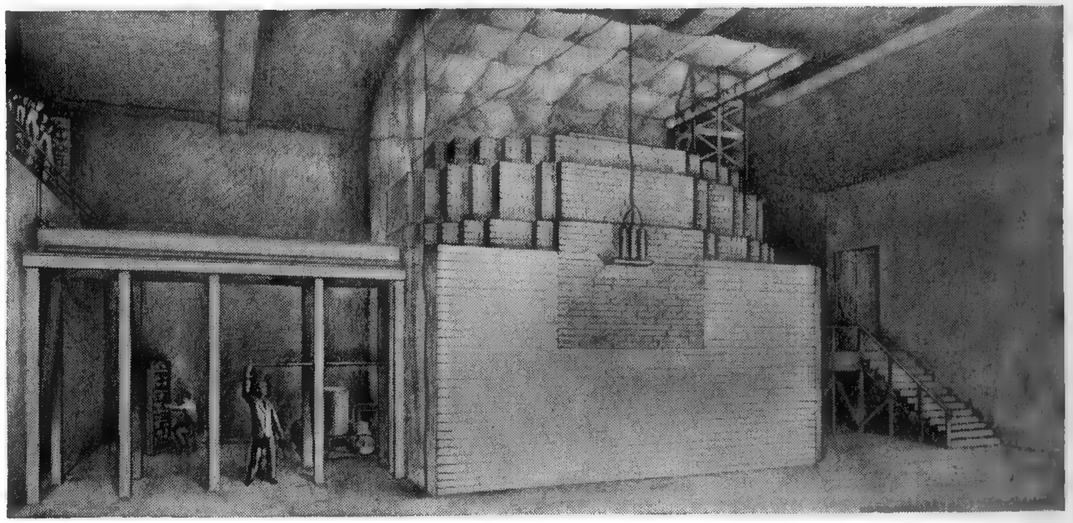


FIG. 13.2 Sketch of completed CP-1 (by Melvin Miller). (Argonne National Laboratory)

at first operated manually, as shown in the drawing in Fig. 13.2.* This drawing also shows the incompleted upper layers and the neutron detecting chambers suspended in front of the reactor. Hand manipulation of the regulating rods was soon replaced by motor control, and an electrically operated solenoid was

* The drawing also shows the balloon cloth (partially rolled up) which could have been used to enclose the reactor. Because of the uncertainty of the critical calculations and the limited availability of uranium, it was thought that the air might have to be replaced with carbon dioxide or helium, in order to eliminate the poisoning effect of nitrogen, with its relatively high cross section for thermal neutrons. However, as events turned out, this proved to be unnecessary.

used in conjunction with an ionization chamber to trip a weight which pulled a safety rod into the reactor in an emergency. CP-1 was generally operated at the low power of 0.5 watt, although the power was increased to 200 watts for a short time. Owing to the absence of a shield, the power had to be kept low to protect operating personnel.

Chicago Pile-2 (CP-2) *

Fuel: 45 tons natural uranium (10 tons as metal, the remainder as oxide, mainly UO_2)
Moderator: 472 tons graphite (including 127 tons reflector)
Reflector: 1 ft graphite
Shield: 5 ft concrete at sides, 4 ft wood and 6 in. lead at top
Over-all dimensions:† 30 × 32 × $25\frac{1}{2}$ ft high
Controls: 3 safety, 1 shim, 2 regulating (cadmium strip on steel)
Coolant: ambient air
Power: usually 2 kw
Thermal flux (av): 3×10^8 neutrons/(cm²)(sec)

13.6. Toward the end of February, 1943, CP-1 was dismantled and the materials, together with additional uranium metal, were used in the construction of

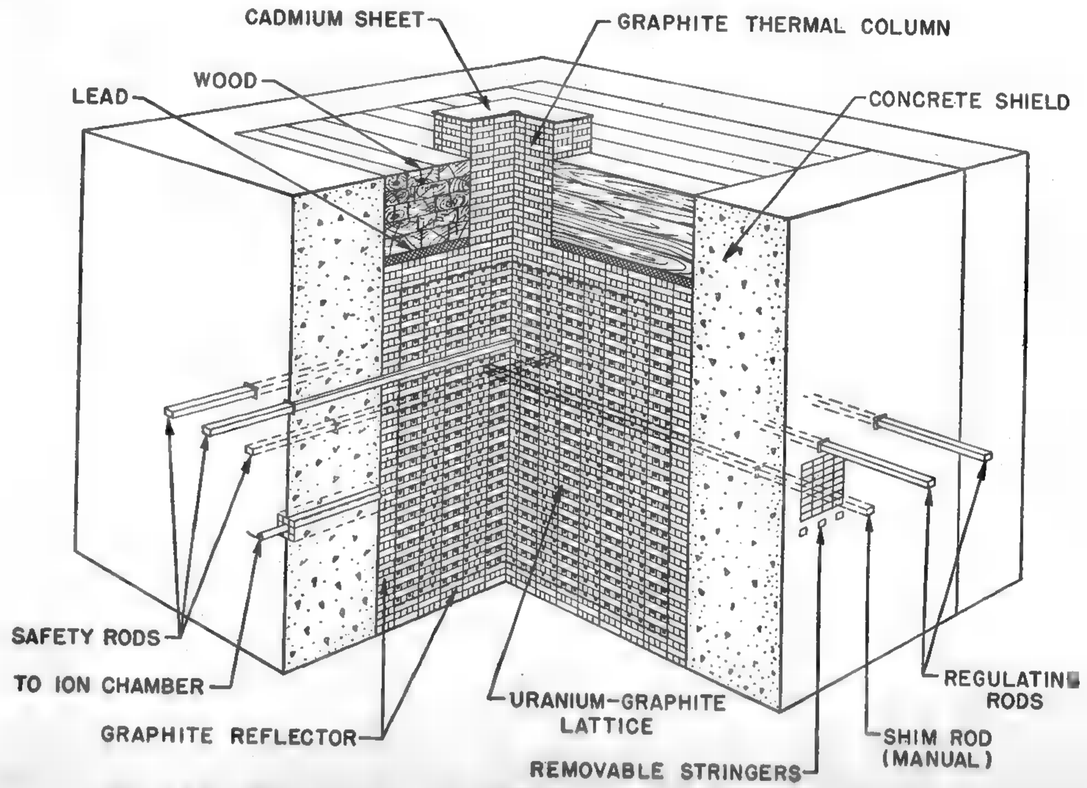


FIG. 13.3 Cutaway drawing of CP-2 at Argonne National Laboratory. (Argonne National Laboratory)

* See references to CP-1, also Report CP-2450.
† The over-all dimensions given here and elsewhere in this chapter include the shield; other dimensions will be found in the text.

CP-2 at the Palos Park site of what is now the Argonne National Laboratory, near Chicago. The active lattice is in the form of a rectangular parallelepiped, approximately 18 ft wide, 20 ft long, and 19 ft high, with a 1-ft graphite reflector and a 4½-to-5-ft shield on all sides; the total weight is 1400 tons (Fig. 13.3). Since heat is removed only by conduction through the shield, the normal operating power is limited to 2 kw, but this has been increased to 100 kw for periods of an hour or more.

13.7. The reactor has a number of experimental facilities, consisting of slots or channels, made by removal of graphite blocks from core and reflector. These can be used for the exposure of materials to neutrons or to obtain beams for experiments outside the reactor. In the top of the reactor is a "thermal column," consisting of a 5 × 5 ft column of graphite blocks, extending from the reactor core through the wood and lead shield, as shown in Fig. 13.3. The top of the thermal column is covered with cadmium; this has an adjustable opening through which neutrons are permitted to escape when required for experimental purposes. These neutrons are completely thermalized, with few, if any, having energies in excess of the thermal value. If the thermal column is long enough (i.e. > 6 ft), and the neutron beam is well collimated, the only neutrons emerging will be those with energies less than 0.0018 ev, the others having been removed by scattering.*

Graphite Low Energy Experimental Pile (GLEEP) †

Fuel: 33 tons natural uranium (13 tons as metal, remainder as UO_2); cylindrical
Moderator: 550 tons graphite (including reflector)
Reflector: 2 ft graphite (octagonal)
Shield: 5 ft concrete (cubic)
Controls: 6 safety, 4 shim, 1 regulating (cadmium)
Coolant: air at 5000 ft³/min
Power: 100 kw
Thermal flux (av): 3×10^{10} neutrons/(cm²)(sec)

13.8. The Graphite Low Energy Experimental Pile (GLEEP) at Harwell, England, is somewhat similar to CP-2, but, having forced air cooling, it can operate normally at 100 kw power. To prevent oxidation of the uranium, at the higher surface temperatures, both the uranium metal and the oxide are canned in aluminum. The cylindrical fuel elements are arranged in lines, rather than in individual lumps as in CP-2, in a square lattice of 1760 channels on 7½-in. centers. The core is in the form of a cylinder about 20 ft in diameter and 17 ft high.

* See S. Glasstone, "Sourcebook on Atomic Energy," D. Van Nostrand Co., Inc., 1950, §§ 11.89, 14.86.

† F. C. W. Colmer and D. J. Littler, *Engineering*, **170**, 497 (1950); *Nucleonics*, **8**, No. 1, 3 (1951); see also, *Chem. Eng. News*, **29**, 113 (1951); *Atomics*, **2**, 51 (1951).

*Oak Ridge Graphite (X-10) Reactor**

Fuel: 54 tons uranium metal
Moderator: 620 tons graphite (including reflector)
Reflector: 2 ft graphite
Shield: 2 ft ordinary concrete + 5 ft special concrete
Over-all dimensions: 38 × 47 × 32 ft high
Controls: 3 safety, 2 shim, 2 regulating
Coolant: air at ~120,000 ft³/min (7250 lb/min)
Power: 3800 kw
Thermal flux (av): 5×10^{11} neutrons/(cm²)(sec)

13.9. The Oak Ridge Graphite Reactor, often referred to as the X-10 reactor because of the map designation of the site, was completed in November, 1943. It was intended to serve to some extent as a pilot plant, bridging the gap between CP-2 and the Hanford reactors, and also to provide gram quantities of plutonium required for testing the chemical separations process for the treatment of spent fuel. However, from the beginning this reactor has been used both as a research tool and for training personnel in reactor operation. Another important function of the X-10 reactor is the production of isotopes by exposure of various elements to neutrons. The reactor was originally designed to operate at 1000 kw, but modifications, including the installation of new fans to provide

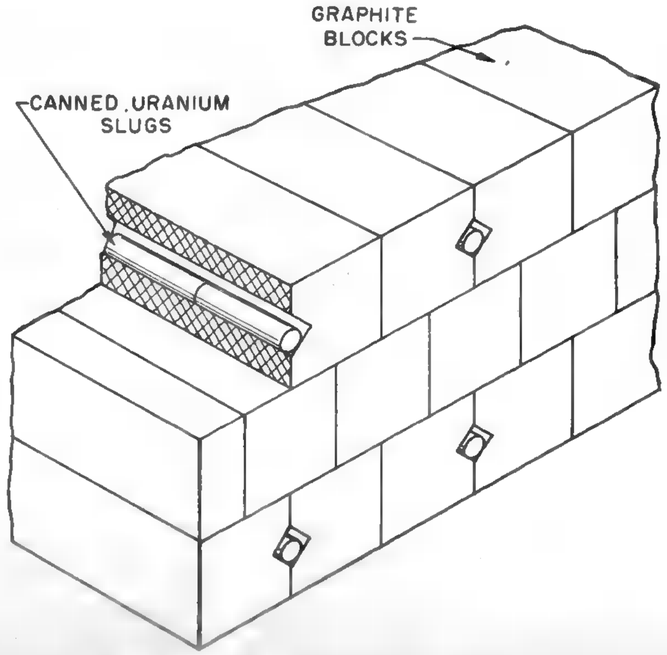


FIG. 13.4 Graphite-uranium lattice

* M. E. Ramsay and C. D. Cagle, *Chem. Eng. Prog. Symposium Series*, No. 11, **50**, 149 (1954); see also *Nucleonics*, **10**, No. 2, 24 (1952).

additional cooling, permitted the power level to be raised to an average of 3800 kw.

13.10. The basic structure of the reactor, including the moderator, is a 24-ft cube of graphite blocks, each being 4 in. square and about 50 in. long. Longitudinal V-shaped cuts are made in adjacent blocks so that they form $1\frac{3}{4}$-in. square channels running right through the reactor (from east to west). There are 1248 of these channels, spaced on 8-in. centers, and in them are inserted the fuel elements (Fig. 13.4). The latter consist of short cylinders or "slugs" of natural uranium metal, 4 in. long and 1.1 in. in diameter, weighing roughly 2.57 lb, with a cladding of 2S aluminum about 0.035 in. thick to protect the uranium from attack and to retain the fission products. The slugs are loaded, end to end, into the reactor, with 39 to 54 slugs per channel, to form what is essentially a long cylinder of fuel. The short slugs are easier to fabricate and are also more convenient to handle in loading and unloading the reactor than long rods.

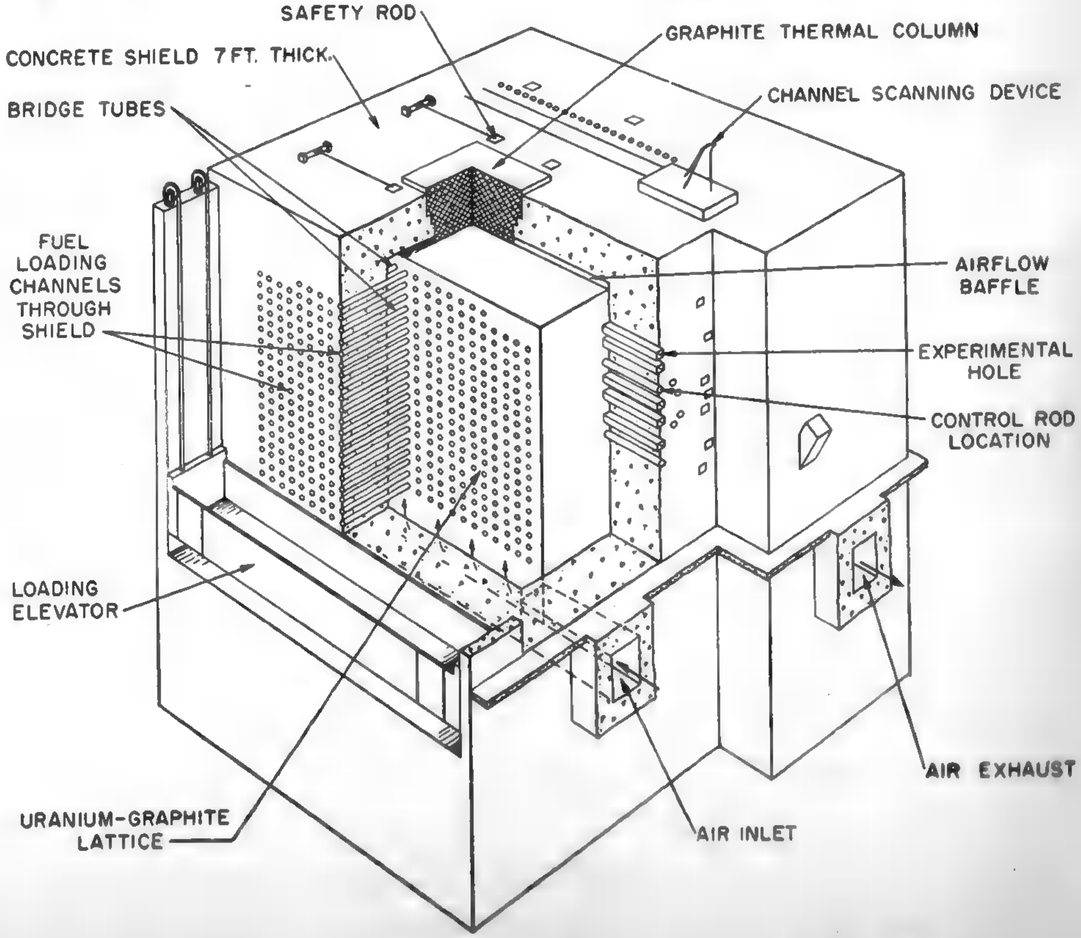


Fig. 13.5 Cutaway drawing of (X-10) natural uranium-graphite reactor at Oak Ridge National Laboratory. (Oak Ridge National Laboratory)

13.11. The graphite cube is separated from the shield by a 3-ft wide air chamber in front (east) and a 6-ft wide chamber at the back (west); as will be seen shortly, these act as plenum chambers for the air cooling the reactor. The shield consists of three layers: the inner and outer layers are each of ordinary (steel reinforced) concrete 1 ft thick, whereas the middle layer, 5 ft thick, is a special concrete mixture containing the clay mineral *haydite*, to retain water, and barytes aggregate to increase the density (§ 10.44). It is doubtful, however, if the haydite serves its intended purpose of improving the neutron absorption properties. Nevertheless, the shield has proved effective in reducing the radiation well below the maximum permissible exposure, even though the operating power is nearly four times the design level.

13.12. The 3-ft wide space between the shield and the front of the graphite cube is bridged by a number of steel tubes, one corresponding to each fuel channel. These pass through the shield and are normally closed by plugs at the reactor (east) face (Fig. 13.5). To load the reactor the shield plugs are removed and the required number of fuel slugs are pushed manually into the core, employing long steel rods. In order to unload the reactor, when shut down, the rods are used to push out the spent fuel plugs at the rear end of the reactor. Here the discharged plugs strike slanted plates and slide into a pool of water 20 ft deep; the water serves as a radiation shield, under which the spent fuel slugs can be manipulated with long-handled tools. By introducing fresh fuel slugs and pushing them along the channels the reactor can be simultaneously unloaded and reloaded. To obviate the danger from radiation the reactor must be shut down when performing these operations. About half an hour is allowed for decay of radioactive aluminum-28 and short-lived, highly active fission products.

13.13. The atmospheric air for cooling the reactor is first drawn, at the rate of 7250 lb/min, through filters to remove dust particles which might become radioactive; it is then passed up the 3-ft plenum chamber at the front of the reactor. From the chamber, the air enters the spaces in the fuel channels around the slugs; slots cut in the bridge tubes, mentioned above, provide additional space for entry of air. After passage along the tubes the heated air is withdrawn from the 6-ft plenum chamber at the rear of the reactor into filters which remove essentially all the radioactive particles present. The exhaust air now enters two 900-hp fans which discharge it to the atmosphere through a 200-ft high stack. By drawing the air through the system in this manner, the interior of the reactor is maintained under reduced pressure, so that there is no danger of radioactive air leaking out into the reactor building.

13.14. To protect the concrete shield at the rear of the reactor from the hot air in the exit plenum chamber, there is a 2-in. layer of an insulating material, Cellamite, separated from the shield by a 4-in. space. The passage of cool air through this space also helps to remove some of the heat arising from the absorption of gamma rays and the slowing down of neutrons in the first few inches of

the concrete. A temperature maximum occurs at a depth of about 1 ft within the shield.

13.15. The three safety rods, which shut the X-10 reactor down quickly in the event of an emergency, consist of cadmium encased in steel. They are suspended vertically, through the top shield, from cables wound around drums held by electric brakes. If safety signals indicate that the reactor should be shut down or if there is a power failure, the brakes are released and the rods fall into the reactor. Normal shut-down is achieved by the same mechanism. As a back-up safety, to be used as a last resort, boron-steel shot, released from containers hanging above the reactor, drops into holes in the core. It is noteworthy that when the X-10 reactor was first operated there were seventeen conditions which caused the reactor to shut down automatically. With increasing experience these have now been reduced to nine without adding to the operational hazards. This is an interesting illustration of the general type of simplification that may be expected in the design of power reactors in the future.

13.16. The two shim rods, which are horizontal and enter the reactor from the north side, are made of steel containing 1.5 per cent of boron. For normal operation these rods are moved in and out of the reactor by electric motor, but to shut the reactor down, either deliberately or automatically in the event of an emergency, they are driven into the reactor, for their full length, by oil pressure. This is achieved by means of a solenoid-operated valve, which is normally closed but is opened by shut-down signals (or by power failure). The hydraulic pressure of the oil is then applied to a piston connected to the rod-drive system and the rod is forced into the reactor core (cf. § 6.86).

13.17. The regulating rods are of boron steel, similar to the shim rods; they are also horizontal and enter the reactor from the north side. These rods for fine control of the power cannot be moved rapidly. They are driven electrically using hydraulic transmission, and their positions are recorded on indicators, calibrated in terms of reactivity, on the control panel. Because changes in reactivity during normal operation are small, automatic control has been found to be unnecessary. The rods are, therefore, moved in or out, as required to maintain the power level constant, by signals supplied manually at the control panel.

13.18. The instrumentation of the X-10 reactor is similar to that described in Chapters V and VI. In addition to the use of ionization chambers and neutron thermopiles for flux measurements, the power level is determined from the product of the rate of flow of air through the reactor and the temperature rise. A special feature is the system of thermocouples, for determining actual temperatures, attached to the cladding of fuel slugs located in positions where the temperature is likely to be high. The operating power level of the reactor is adjusted so that in no case does the observed temperature of the fuel cladding exceed 245°C (473°F).

13.19. The X-10 reactor has numerous openings, channels, and beam holes,

of several different sizes, that can be used for a variety of experiments with neutrons. The Lid Tank Shielding Facility, described in § 10.121, is located at the west face of the reactor. There is also a 5 × 5 ft thermal column which projects through the top shield (see Fig. 13.5).

*The Brookhaven (BNL) Reactor**

Fuel: 60 tons natural uranium metal
Moderator: 700 tons graphite (including reflector)
Reflector: 3½ ft graphite
Shield: 9–18 in. steel + limonite-iron concrete up to 5 ft total
Over-all dimensions: 38 × 55 × 30 ft high
Controls: 14 shim-safety, 2 regulating
Coolant: air at 270,000 ft³/min
Power: 28,000 kw
Thermal flux (av): $\sim 3 \times 10^{12}$ neutrons/(cm²)(sec)

13.20. The Brookhaven National Laboratory (BNL) reactor, completed in August, 1950, was designed for research purposes with isotope production as a secondary objective. Although the neutron flux attainable in the reactor is not as high as in some other research reactors, e.g., NRX, MTR, it has more space for experimental facilities because of its larger size (Fig. 13.6).

13.21. The general design of the BNL reactor is similar to that of the Oak Ridge graphite reactor. The core and reflector are in the form of a 25-ft cube made of graphite blocks; this cube is pierced by 1369 circular fuel channels, in a square array on 8-in. centers, in which the aluminum-clad fuel slugs are placed. The shield is of special iron-containing concrete having a density of 4.27 g/cm³; it is enclosed in forms of 3-in. steel plate and so, with an additional 3 in. of steel around the reactor, there is an effective thermal shield 6 in. thick.

13.22. Probably the most important design difference between the BNL and X-10 reactors lies in the cooling system. The active cube of the former is split vertically into two halves, with a gap of 2 to 3 in. between them. The filtered cooling air from the atmosphere is introduced here, where the neutron flux is highest, and then divides into roughly equal parts; the air then flows through the cooling channels toward the outer face in either half of the core (Fig. 13.7). Although the total mass rate of flow of air is less than three times that of the Oak Ridge X-10 reactor, the normal power level, which this cooling arrangement makes possible, is about six times as great. After passage through the fuel channels the air enters plenum chambers, 14 ft wide at the front of the reactor and 6 ft wide at the back. From these chambers the exhaust air is sucked through filters and is cooled before being discharged by fans to the 320-ft-high stack. The temperature of the air leaving the reactor at its full operating power is about 330°F and plans were made to use this heat for warming buildings. However, since a conventional standby heating plant was required, in any

* L. B. Borst, *Physics Today*, **4**, 6 (1951); Report AECU-1089; see also *Chem. Eng.* **58**, No. 5, 271 (1951).

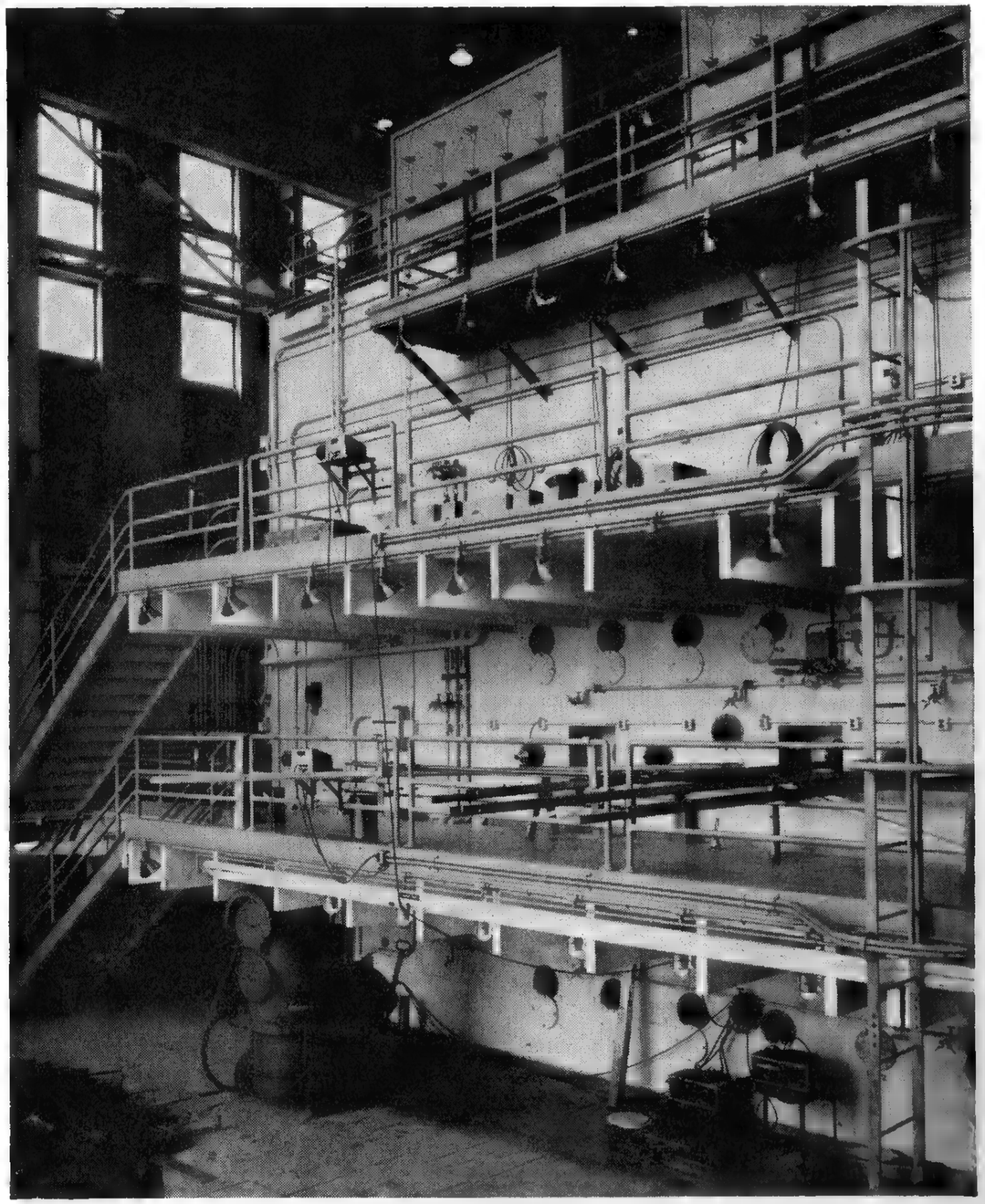


FIG. 13.6 West face of reactor at Brookhaven National Laboratory. (Brookhaven National Laboratory)

event, when the reactor was shut down, the scheme for using the nuclear heat was abandoned.

13.23. In order to keep as much as possible of the vertical faces and the top of the reactor available for experiments, the boron-steel control rods are all horizontal; they enter the reactor diagonally, eight from each of two corners. Each of the 14 emergency shim-safety rods is moved by its individual electric-motor-driven hydraulic system. If the main power fails, the energy stored in

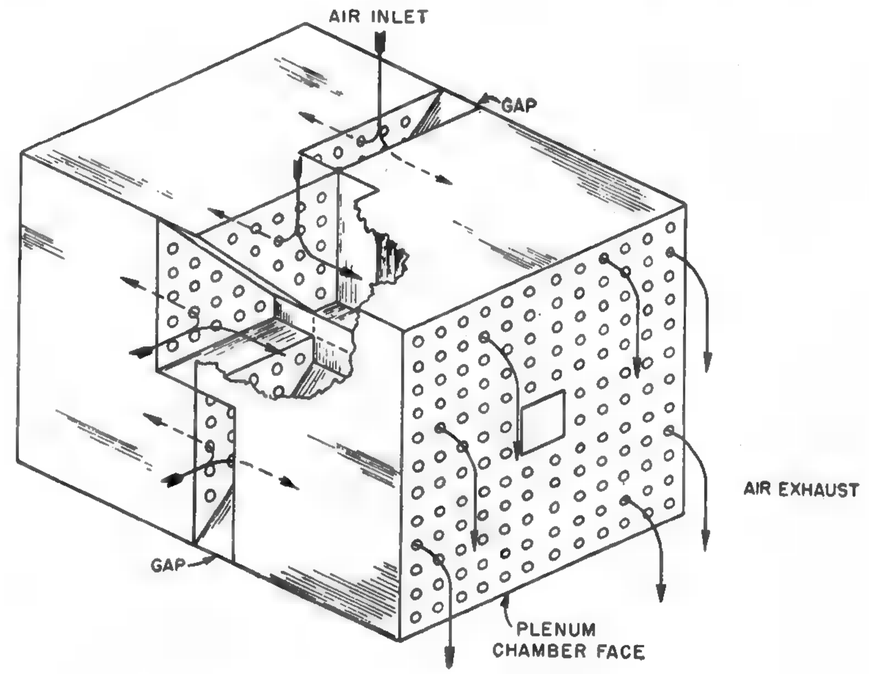


FIG. 13.7 Flow of air through Brookhaven reactor. (Brookhaven National Laboratory)

the fly wheel on the shaft of each electric motor is sufficient to drive the emergency rods into the reactor core. The two regulating rods are moved by an induction motor which is subject to automatic control once the reactor has been brought up to the desired power level. A manually controlled, back-up safety arrangement of boron-steel shot is available to shut down the reactor in the last resort.*

13.24. In addition to the difference in the cooling system, the BNL and Oak Ridge natural-uranium reactors also differ in the method of fuel loading and unloading. The BNL reactor does not have bridge tubes across the front plenum chamber, and loading is achieved by placing removable troughs across the air gap. The spent fuel plugs are also withdrawn from the front of the reactor into the bridge troughs. From here the slugs are positioned by a remote-control device and lowered into a 20-ft deep pool of water.

* For details of the control panel of the BNL reactor, see Fig. 6.7.

13.25. Of the many experimental facilities available only a few need be chosen for special mention. Among these are the two large tunnels under the reactor which permit large animals for experiments or instruments to be located near the core. An endless chain of sample holders, extending through the reactor and the shield, permits the removal of exposed materials, e.g., radioisotopes, without shutting down the reactor. When a larger area of irradiation surface is required, a 20-ft square section of the shield at the top of the reactor can be removed. The BNL reactor is also equipped for shielding design studies.

British Experimental Pile (BEPO) *

Fuel: 40 tons natural uranium metal (28 tons for criticality)
Moderator: 600 tons graphite (including reflector)
Reflector: about 3 ft graphite
Shield: 6 in. cast iron + 6 ft barytes concrete
Controls: 10 vertical + 4 horizontal (steel tubes containing boron carbide)
Coolant: air at 260,000 ft^3/min
Power: 6000 kw
Thermal flux (av): $\sim 1 \times 10^{12}$ neutrons/$(cm^2)(sec)$

13.26. The British Experimental Pile (BEPO), at Harwell, England, used for research and radioisotope production, resembles the Oak Ridge X-10 reactor, although it has a larger cooling capacity and, hence, operates at a higher power level. The core of the BEPO reactor is in the form of a right cylinder about 20 ft in diameter and 20 ft high. The cylindrical fuel rods, 0.9 in. in diameter and 12 in. long, are loaded into channels, about 20 to a channel, which are spaced $7\frac{1}{4}$ in. apart in a square array. There are 1400 fuel channels running through the graphite matrix, but only about half of them, in the central region, are used in normal loading. The maximum permissible surface temperature of the aluminum-clad fuel rods (in cartridges) is 200°C (390°F). The heat in the exhaust air, which leaves the reactor at roughly 175°F, is used for warming buildings.

Sodium-Graphite Reactor†

13.27. It was seen in Chapter XII that a reactor moderated by graphite and cooled with sodium has much to recommend it for nuclear power production. These materials have the advantage of permitting reactor operation at high temperatures, so that high thermal efficiencies for electrical power generation are possible.

13.28. An experimental reactor of this type, called the Sodium Reactor Experiment (SRE), is included in the A.E.C.'s development program.* The fuel rods of slightly enriched uranium, clad with stainless steel, will be held vertically in channels through which the liquid sodium flows. The core will consist of a number of cells containing graphite, enclosed in a metal (stainless steel or zirconium) can, with a fuel and coolant channel at the center. The assembly of cells will completely fill a steel tank capable of withstanding the corrosion of liquid sodium at high temperature. The liquid sodium will enter the tank at about 500°F and will leave somewhere from 900° to 1000°F. The heat output of the SRE will be 20,000 kw; the design will resemble that of a full-scale power plant and there will be equipment for generating electricity.†

13.29. With the slightly enriched uranium fuel to be used, initially at least, a conversion ratio (to plutonium-239) of 0.9 is to be expected. The same system may later be operated as a thermal power-breeder with uranium-233 as the fissionable species and thorium as the fertile material. Among the studies to be made with the SRE are investigations of fuel element, coolant, and structure temperatures, fuel burn-up, corrosion, etc.

Heavy-Water-moderated Reactors: Natural Uranium Fuel

Chicago Pile-3 (CP-3) ‡

Fuel: 3 tons natural uranium metal
Moderator: $6\frac{1}{2}$ tons heavy water (in cylindrical tank)
Reflector: 2 ft graphite; heavy water at top
Shield: 4 in. Pb-Cd + $7\frac{3}{8}$ ft concrete (octagonal)
Over-all dimensions: octagonal, 26 ft across × 14 ft high
Controls: 2 safety, 3 shim, 2 regulating
Coolant: circulation of heavy-water moderator (200 gal/min)
Power: 300 kw
Thermal flux (av): 5×10^{11} neutrons/$(cm^2)(sec)$

13.30. With the increasing availability of heavy water, the construction of a reactor using this material as moderator became possible, and the first such reactor, the CP-3, was completed in May, 1944, at the Argonne National Laboratory. The reactor was dismantled in January, 1950, because of suspected corrosion of the aluminum cladding of some of the fuel rods, and the natural uranium fuel was replaced with enriched fuel. The new reactor was named CP-3 prime (CP-3′) and this will be described later. Although CP-3 is no longer in existence as such, a description is of interest because its design was somewhat similar to other natural uranium-heavy water reactors of more recent construction, e.g., the Norwegian-Dutch (JEEP), and the Swedish reactors.

* *Nucleonics*, **8**, No. 6, 36 (1951); *Atomics*, **2**, 176 (1951); **5**, 115 (1954); J. Cockcroft, *Proc. Inst. Elec. Eng.* (*Lond.*), Part I, **100**, 83 (1953).

† C. Starr, Atomic Industrial Forum Report, July 1954, p. 11; W. E. Parkins, *Chem. Eng. Prog. Symposium Series*, No. 13, **50**, 53 (1954); see also "Report to the U. S. Atomic Energy Commission on Nuclear Power Technology," U. S. Government Printing Office, May 1953, p. 12; *Nucleonics*, **11**, No. 6, 62 (1953).

* "Report on the Five-Year Power Reactor Development Program Proposed by the Atomic Energy Commission," U. S. Government Printing Office, March 1954, p. 12.

† *Nucleonics*, **13**, No. 5, 14 (1955).

‡ Report AECD-3415; W. H. McCorkle, *Nucleonics*, **11**, No. 5, 21 (1953); S. McLain, *Proc. Kjeller Conf. on Heavy Water Reactors*, 1953, Joner Publications No. 7, p. 189.

In any event, the reflector, shield, controls, and cooling system were not altered when the fuel elements of CP-3 were changed.

13.31. The core of the CP-3 reactor was contained in a cylindrical aluminum tank, 6 ft in diameter and nearly 9 ft high, in which some 120 (or more) aluminum-clad rods of natural uranium metal were suspended vertically in 6½ tons of heavy water. The rods were cylindrical, 1.1 in. in diameter and 6 ft long,

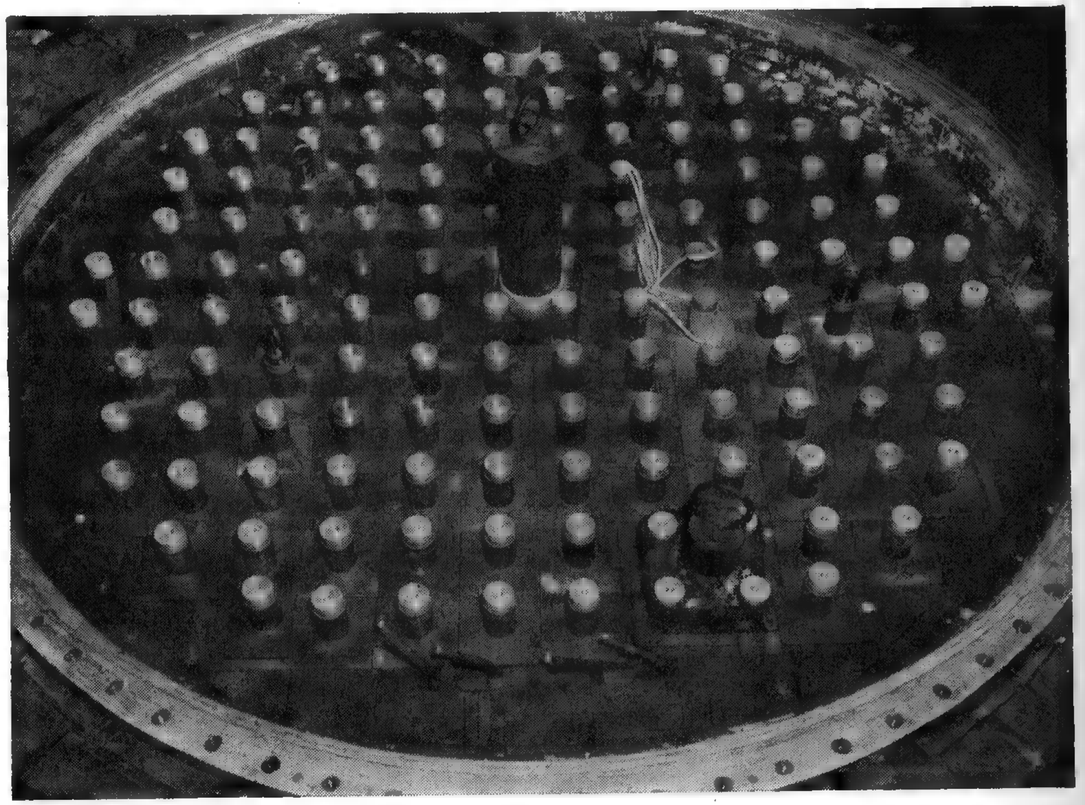


FIG. 13.8 Top of core tank of CP-3 at Argonne National Laboratory, showing fuel rods. (Argonne National Laboratory)

arranged in a square lattice on 5⅜-in. centers (see Fig. 13.8). The heavy water was circulated through an external heat exchanger, at a rate of about 200 gal/min, where it was cooled with ordinary water, before being returned to the reactor at 87°F.

13.32. The core tank of the reactor is surrounded at the bottom and sides by a 2-ft layer of graphite to act as reflector; heavy water covering the fuel rods acts as the reflector at the top of the reactor. The thermal shield of 4-in. thick cadmium-lead alloy is cooled with water flowing through copper tubes. The biological shield, of octagonal shape, consists of 7⅔ ft of concrete at the sides; on the top there is a layer of cadmium, 1 ft of lead, and 4 ft of alternate layers of

steel and compressed wood fiber (Masonite) in the form of removable blocks (Fig. 13.9).

13.33. The control rods consist of a layer of cadmium in the annular space between inner and outer aluminum cylinders. An unusual feature of the control system is that one of the safety rods and one regulating rod are of the signal-arm type, hinged to the bottom of the reactor cover-plate near the edge. For normal reactor operation the rods are held in a horizontal position, but for emergency or intentional shut-down or in the event of a power failure the rods

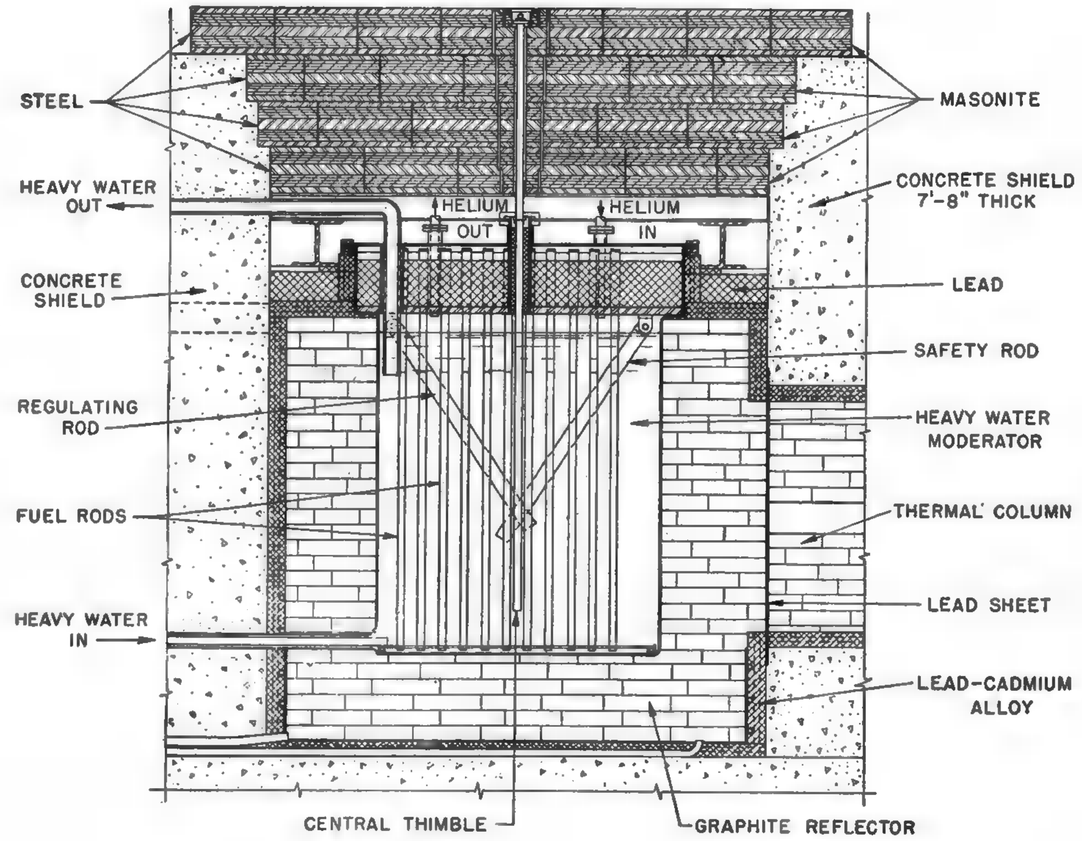


FIG. 13.9 Sectional drawing of CP-3 (and CP-3′) reactor. (Argonne National Laboratory)

drop into the reactor core. As a final safety measure the heavy water moderator can be dumped to a storage tank through a large, quick-opening valve.

13.34. In order to prevent atmospheric moisture from coming into contact with the heavy water, the space in the cylindrical core-tank above the moderator is filled with helium. By circulating this purge gas, the small quantities of deuterium and oxygen gases formed by radiation decomposition of the heavy water are continuously removed and transferred to a catalytic recombiner, thus

obviating a possible explosion hazard (§ 8.148). The heavy water formed in the recombiner is returned to the reactor. The elimination of atmospheric nitrogen from the space above the reactor core also prevents the formation, under the influence of radiation, of nitric acid, which would corrode the aluminum tank and fuel cladding.

13.35. In addition to the usual experimental facilities, such as thermal column, beam openings, and channels into the reflector and shield, the CP-3 reactor has what is known as a central experimental thimble, consisting of a 4-in. diameter aluminum tube, extending into the core (Fig. 13.9). In this tube it is possible to expose materials to the maximum neutron flux, which exists at the center of the reactor.

*NRX Reactor**

Fuel: ~10.5 tons natural uranium metal
Moderator: ~17 tons heavy water
Reflector: graphite

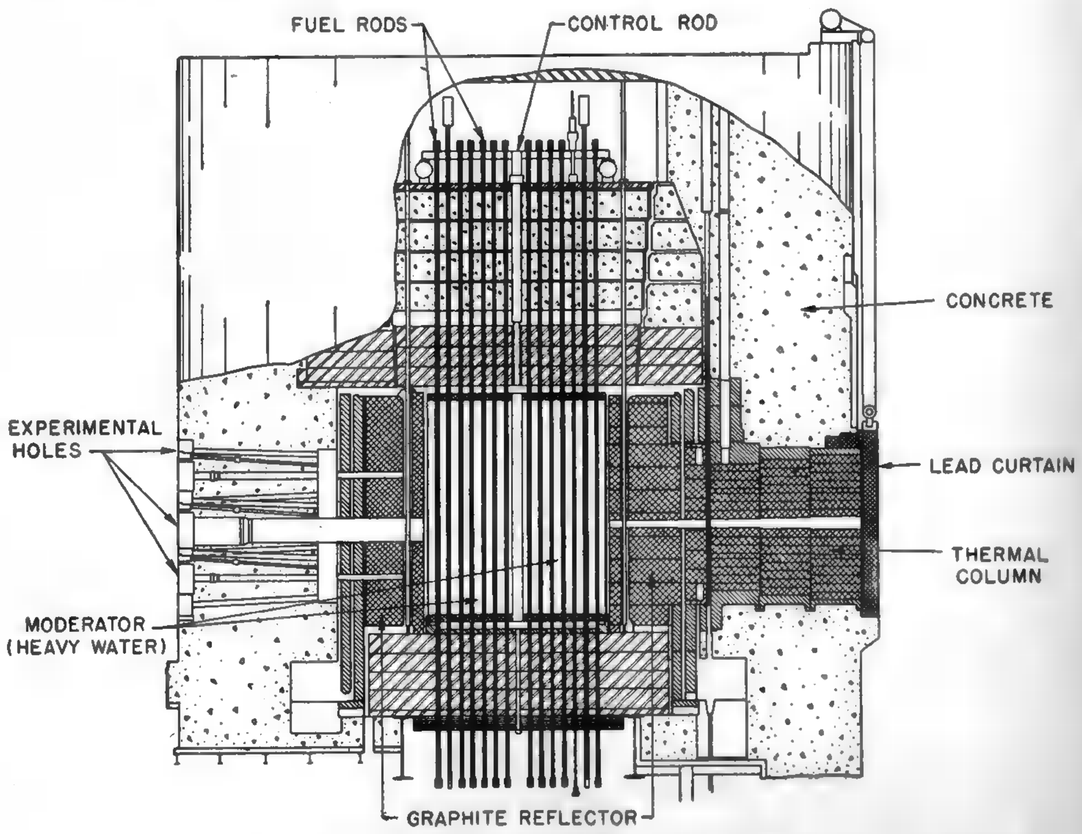


FIG. 13.10 Cross section of Canadian NRX reactor at Chalk River, Ontario. (National Research Council, Canada)

* W. B. Lewis, *Physics Today*, **4**, No. 11, 12 (1951); D. G. Hurst, *Elec. Eng.*, **70**, 476 (1951); F. W. Gilbert, *Nucleonics*, **10**, No. 1, 6 (1952); *Chem. Eng. Prog.*, **50**, 267 (1954); C. R. McCullough, Report AECU-2900, p. 25.

Shield: steel (thermal shield) + 8 ft concrete
Over-all dimensions: cylinder, 34 ft diameter × 34 ft high
Control: vertical rods and variation of moderator level
Coolant: ordinary water
Power: 30,000 kw
Thermal flux (av): 2×10^{13} neutrons/(cm²)(sec)

13.36. For some time after its completion in August, 1947, until the MTR commenced operation, the Canadian National Research (Council's) Experimental (NRX) reactor at Chalk River, Ontario, had the highest thermal neutron flux of any known reactor. The heavy-water moderator is contained in an aluminum tank, called a calandria, 8 ft diameter and 10 ft high. In this are supported the 176 vertical fuel rods of natural uranium, as indicated in Fig. 13.10. The design of the cooling system will be understood from Fig. 13.11, which shows a section through an aluminum-clad fuel rod, surrounded by two concentric aluminum tubes. In the inner annulus the cooling water flows downward, while in the outer annulus a stream of air flows upward. One of the purposes of the air gap is to prevent contamination of the heavy water by ordinary water in the event of a leak. The moderator is also cooled by circulation through a heat exchanger outside the reactor. The helium purge system is similar to that of the CP-3, described above.

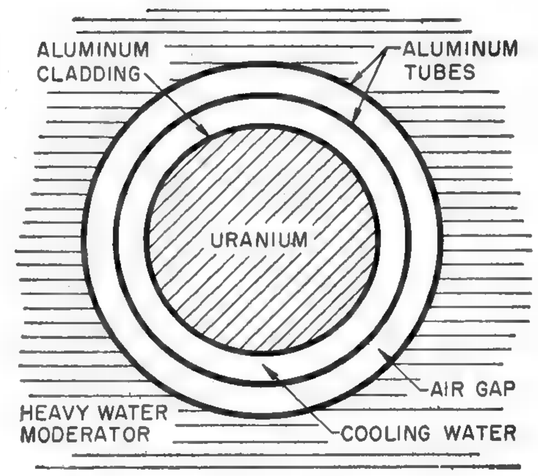


FIG. 13.11 Fuel rod and coolant channel of NRX reactor

13.37. The NRX reactor is controlled by vertical steel rods containing either cadmium or boron; rapid dumping of the moderator for emergency shut-down is possible, as with CP-3. An interesting feature of the control system is that coarse (or shim) control is achieved by varying the level of the heavy water moderator. That the reactor has many research facilities will be evident from the photograph in Fig. 13.12.

13.38. Owing to an unusual combination of accidental circumstances, in December, 1952, a number of fuel rods of the NRX were ruptured so that there was considerable radioactive contamination of the cooling-water system and the reactor structure. It was at first thought that the reactor would have to be abandoned, but as the result of a determined effort, it was decontaminated and reconstructed. The restored NRX reactor commenced operation in February, 1954.

13.39. Another experimental reactor, called the NRU reactor, is being built

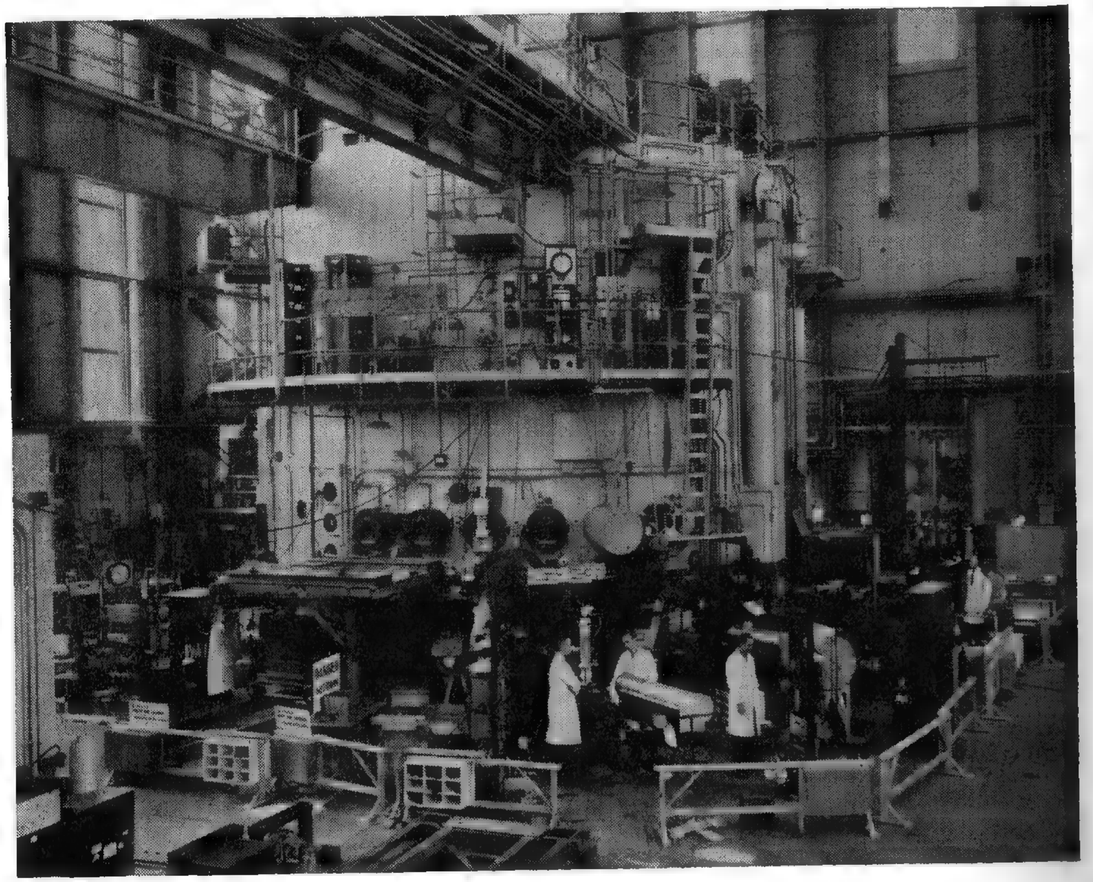


FIG. 13.12 General view of NRX reactor. (National Research Council, Canada)

in Canada. It is to be a natural uranium-heavy water system, like the NRX, but its power is to be higher.

OTHER HEAVY WATER-NATURAL URANIUM REACTORS

13.40. The Zero Energy Experimental Pile (ZEEP),* the first to be built in Canada, was completed in August, 1945. The fuel is uranium metal in the form of short cylinders stacked in vertical aluminum tubes. The latter are suspended in a cylindrical steel tank ($5\frac{3}{4}$ ft in diameter $\times$ $8\frac{1}{2}$ ft high) containing the heavy-water moderator. The reflector is of graphite. There is no cooling and only limited shielding, so that the normal operating level is kept down to 3.5 watts. The average thermal flux is about 6×10^6 neutrons/(cm^2)(sec). A somewhat similar reactor is the Deuterium Moderated Pile, Low Energy (DIMPLE), completed at Harwell, England, in July 1954. The vertical fuel rods are immersed in the heavy-water moderator contained in an aluminum tank; this is surrounded by a graphite reflector and a concrete radiation shield. In effect DIMPLE is really a versatile critical assembly of essentially zero power (cf.

* B. W. Sargent, Report PD-207 (or NRC-1685); *Atomics*, 1, No. 14, 22 (1950).

§ 12.108) in which both the nature of the fuel and its arrangement in the moderator can be readily changed so that a wide variety of designs can be investigated. One function of the assembly is to obtain information for the design of a high-flux reactor for experimental and research purposes.

13.41. Another natural uranium-heavy water reactor of very low power is the one in Chatillon, France, sometimes called ZOE,* from the initial letters of the words Zéro, Oxyde de urane, Eau lourde, representing its power and composition. This reactor, which first became critical in December, 1948, uses uranium dioxide (UO_2) as fuel to save the time required to develop the technique for the production of uranium metal. Pellets of the oxide are packed into aluminum tubes (2.6 in. internal diameter) to form the fuel elements; these are suspended vertically in a cylindrical tank of heavy water. The fuel elements, up to 69 in number, are arranged uniformly in a centered-hexagonal lattice, with a spacing of about $7\frac{1}{3}$ in. At critical loading the core contains about 3.4 tons of oxide (61 tubes), i.e., approximately 3 tons of uranium, and 4.6 tons of heavy-water moderator; an additional amount of heavy water at the bottom of the tank serves as reflector. The main reflector surrounding the tank is a 3-ft thick layer of graphite blocks. The reactor has a 5-ft concrete shield, but it has no forced cooling and so the normal power level does not exceed 5 kw; the average thermal flux is then about 10^{10} neutrons/(cm^2)(sec).

13.42. The second French reactor (P-2), located at Saclay,† was started up in October, 1952. It is unique in being the only known heavy-water reactor to employ a gas for forced internal cooling. One reason for this type of cooling, rather than circulation of the moderator, as in most heavy-water reactors, was that it seemed to be of special interest for power production. The reactor has 136 aluminum-clad vertical fuel rods of uranium metal, each of which is 1.1 in. in diameter and about 7 ft long. The total weight of uranium is 3.3 tons and the critical filling of heavy water requires about 6.3 tons, contained in a cylindrical tank $6\frac{2}{3}$ ft in diameter and $8\frac{2}{3}$ ft high. The graphite reflector has a thickness of 3 ft and the whole is surrounded by a cast-iron thermal shield and a biological shield of concrete.

13.43. For cooling purposes each fuel rod is surrounded by two concentric aluminum tubes; coolant gas, usually nitrogen at 10-atm pressure, flows down the outer annulus and then up the inner one. The gas leaving the reactor passes through a water-cooled heat exchanger to a blower, but since it is heated somewhat, due to overcompression, it is sent through another heat exchanger before returning to the reactor. The reflector and shield are cooled by air which is discharged to the atmosphere through a stack.

13.44. When the reactor operates at 1000 kw, which is the limit set by the

* M. E. Nahmias, Report NP-2038; L. Kowarski, *J. phys. radium*, 12, 751 (1951).

† L. Kowarski, *Proc. Kjeller Conf. on Heavy Water Reactors*, 1953, Jener Publications No. 7, p. 144; *Nucleonics*, 12, No. 8, 13, 84 (1954); M. Goldschmidt and F. Perrin, *Chem. Eng. Prog. Symposium Series*, No. 12, 50, 243 (1954).

maximum permissible surface temperature (250°F) of the fuel elements, the compressed nitrogen gas enters the reactor at 77°F and leaves at about 178°F. The average value of the thermal flux at this power level is 2×10^{12} neutrons/(cm²)(sec). In July, 1954 the maximum power of the reactor was increased to 1500 kw by the use of new fuel elements which can withstand higher temperatures.

13.45. Nitrogen was chosen as the reactor coolant because of its chemical inertness and on account of the weak activation by neutrons as compared to air in which the argon becomes highly radioactive (cf. § 9.126). When nitrogen captures neutrons the product is generally carbon-14 and this emits only low-energy beta particles with no gamma radiation. Carbon dioxide has been employed as coolant in the Saclay reactor in place of nitrogen. The former gas has the advantage of smaller neutron absorption and of better heat-transfer properties, but owing to its higher density the pumping power, for the same pressure difference, is greater than for nitrogen. Consideration is being given to the installation of blowers of higher power to permit the use of carbon dioxide as coolant at 10 atm pressure.

13.46. The JENER or Joint (Norwegian-Dutch) Establishment (for Nuclear Energy Research) Experimental Pile, often referred to as JEEP,* at Kjeller, Norway, was first operated in 1951. It was designed independently of CP-3, and its resemblance to the latter implies a similar solution to a particular problem. The main difference between the two reactors is that in the JEEP the fuel rods are arranged in pairs, giving a possible increase in the multiplication factor (cf. § 12.161), whereas in CP-3 they are single.

13.47. The aluminum-clad, vertical fuel rods of the JEEP are 1.0 in. in diameter and have a total length of $6\frac{1}{4}$ ft. There are 150 such rods, consisting of 75 double fuel units, arranged in a lattice having a pitch of slightly more than 7 in. Each unit is made up of two rods rigidly connected with their centers 1.57 in. apart. The total weight of uranium metal in the reactor is only 2.42 tons. The heavy-water moderator (99.8 to 99.85 per cent purity) weighs somewhat over 7 tons and is contained in a tank of $6\frac{1}{2}$ ft diameter. This is surrounded by a 2.3-ft thick layer of graphite to act as reflector. The moderator is circulated at the rate of 60 gal/min through an external water-cooled heat exchanger. Instead of the helium purge system, as used in CP-3, air is passed over the surface of the moderator in the reactor tank and through a catalytic recombiner. The normal operating power of JEEP is 250 to 300 kw and the thermal flux is roughly 10^{12} neutrons/(cm²)(sec).

13.48. The Swedish reactor, built in Stockholm in 1954, was designed to operate at 300 kw, partly on the basis of information available concerning CP-3 and JEEP.† It is, therefore, not essentially different from either of these

* O. Dahl and G. Randers, *Nucleonics*, **9**, No. 5, 5 (1951); T. J. Borendregt and K. H. Brakstad, *Chem. Eng. Prog. Symposium Series*, No. 11, **50**, 183 (1954).

† S. Eklund, *Proc. Kjeller Conf. on Heavy Water Reactors*, 1953, Jener Publications No. 7, p. 127; *Chem. Eng. Prog. Symposium Series*, No. 11, **50**, 189 (1954).

reactors. Like CP-3, the Swedish reactor uses helium as the circulating purge gas above the moderator. The only unusual feature of the reactor is the external heat exchanger in which the heavy-water moderator is cooled by air instead of ordinary water. This was done in order to avoid the possibility of contamination of the valuable heavy water by light water in the event of a leak developing in the heat exchanger.

13.49. The Swedish reactor is the first, as far as is known, to be built in an underground chamber, some 60 ft below surface level. The location was determined by the desirability of having the reactor close to existing laboratories in a part of Stockholm where no other space was available. It was realized, too, that an underground site would provide a measure of protection to the city's population in the event of an accident to the reactor.

Heavy-Water-moderated Reactors: Enriched Uranium Fuel

Chicago Pile-3′ (CP-3′) *

Fuel: 4 kg (9 lb) of uranium-235 (in aluminum alloy)
Power: 300 kw
Thermal flux (av): 2×10^{12} neutrons/(cm²)(sec).

13.50. After the suspected corrosion of some of the fuel rods in CP-3 in January, 1950, it was decided to replace all the natural uranium by enriched material. This made possible an increase in the neutron flux by a factor of four for the same operating power and cooling system. The fuel rods for the modified (CP-3′) reactor are made of an aluminum alloy containing 2 per cent highly enriched uranium; they are 0.85 in. in diameter and $5\frac{1}{2}$ ft long. All the other features of the reactor, which commenced operation in July, 1950, are essentially the same as those of CP-3, already described.

Chicago Pile-5 (CP-5) †

Fuel: 1.2 kg (2.6 lb) uranium-235 (as aluminum alloy)
Moderator: 6.5 tons heavy water
Reflector: 2 ft heavy water + 2 ft graphite
Shield: $\frac{1}{4}$ in. boral, $3\frac{1}{2}$ in. lead, $4\frac{2}{3}$ ft limonite-iron concrete (density 4.4 g/cm³)
Over-all dimensions: octagonal, 20 ft across × $13\frac{1}{2}$ ft high
Control: 4 shim-safety (signal-arm type), 1 regulating (vertical)
Coolant: circulation of heavy-water moderator (~1000 gal/min)
Power: 1000 kw
Thermal flux (av): 2×10^{13} neutrons/(cm²)(sec)

13.51. The CP-5 reactor, which commenced operation in 1954, was constructed as an elaborate research tool for the Argonne National Laboratory to replace CP-2 and CP-3′, which must be dismantled so that the site may be

* See references for CP-3.

† J. M. West, *Proc. Kjeller Conf. on Heavy Water Reactors*, 1953, Jener Publications No. 7, p. 21; S. Untermyer, *Nucleonics*, **12**, No. 1, 12 (1954); J. T. Wellis, *Chem. Eng. Prog. Symposium Series*, No. 11, **50**, 213 (1954).

returned to the owners. Heavy water was chosen as moderator, rather than ordinary water, as in the MTR, because, with a given amount of uranium-235, the former permits the design of a larger core and so provides somewhat more flexibility for experiments.

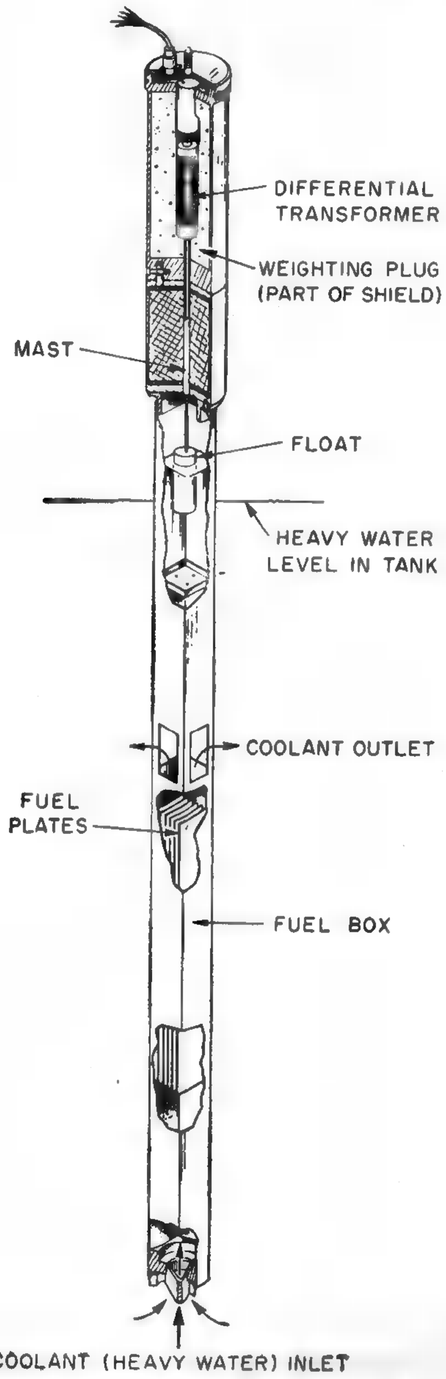


FIG. 13.13 Fuel assembly for CP-5 at Argonne National Laboratory. (Argonne National Laboratory)

13.52. The aluminum tank of CP-5, containing the heavy water, has a diameter of 6 ft and a height of about $7\frac{1}{2}$ ft. The actual fuel-bearing core occupies an approximately cylindrical volume, about 2 ft in diameter and 2 ft high, at the center of the tank. The fuel elements are of the curved sandwich-plate type (cf. § 12.163), made up of a 0.02-in. thick central layer of enriched uranium-aluminum alloy clad on each side with 0.02 in. of aluminum. A fuel assembly consists of ten such plates, 2 ft in length, mounted in a long rectangular box, 3 in. $\times$ 3 in. cross section (Fig. 13.13). For normal operation about twelve of these assemblies, i.e., 120 plates are used; however, there is accommodation for seventeen assemblies, so that additional fuel can be added if required.

13.53. To cool the reactor, the heavy water which leaves the tank at about 126°F is circulated through the external heat exchanger and returns to the reactor at about 118°F. The mass-rate of flow of the heavy water is determined with an electromagnetic flow meter and the difference between inlet and outlet temperature is indicated by electrical resistance thermometers. The product of these quantities is recorded automatically (cf. § 5.128) as the reactor power level on an indicator in the control room.

13.54. In the reactor tank the cooled moderator enters each fuel box through a balancing orifice at the bottom, then flows upward past the fuel plates and leaves through openings just above the plates, as

shown in Fig. 13.13. The arrangement of float, mast, and differential transformer at the top of the fuel box serves the purpose of recording the rate of flow of moderator-coolant. The weighting plug, which helps to keep the fuel box in place, is also part of the reactor shield.

13.55. For control purposes there are four shim-safety rods, made of cadmium clad in aluminum; these are of the signal-arm type, as used in CP-3, and they operate between parallel rows of fuel assemblies. The regulating rod is vertical and is located just outside the reactor core; it can be used to control the reactor power automatically at any desired level. A dump valve permits a rapid lowering of the moderator level if an emergency should arise.

13.56. An unusual feature of CP-5 is reactivity control by cooling the moderator with chilled water at 5°C (41°F). Instead of using extra fuel, in the form of built-in excess reactivity, to allow for xenon poisoning and other transient

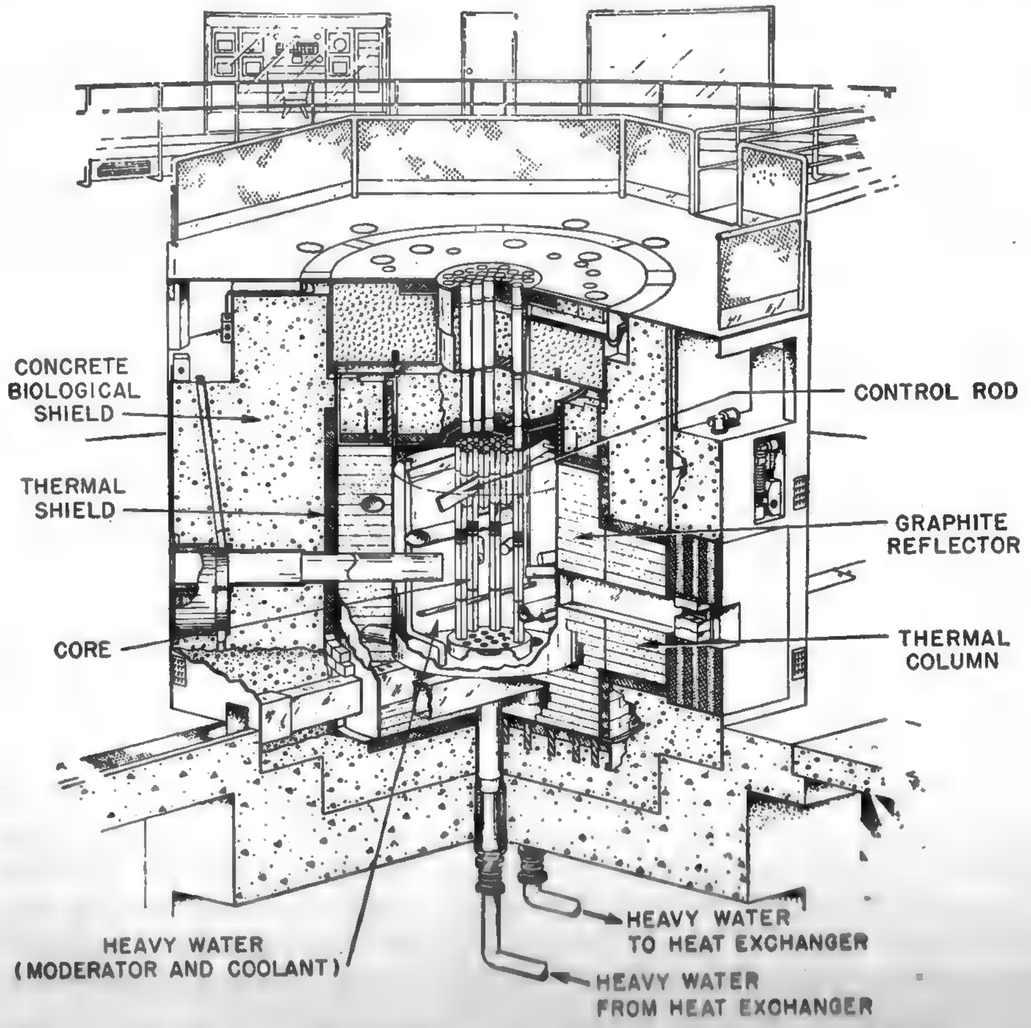


FIG. 13.14 Cutaway drawing of CP-5 reactor. (Argonne National Laboratory)

effects, advantage is taken of an increase of reactivity, resulting from a decrease in the temperature of the moderator, to provide temporary additional reactivity. For example, in starting up after a short shut-down, the additional reactivity required to overcome the accumulated xenon is made available by cooling the moderator with the chilled water. Then, as the excess xenon is consumed, during reactor operation, the moderator temperature is allowed to rise.

13.57. In addition to the helium purge system such as is used in CP-3, a circulating helium atmosphere is employed to cool the graphite reflector. The 3½-in. thick thermal shield of lead, surrounding the graphite zone, is cooled by means of coils through which water flows.

13.58. In accordance with its primary purpose, CP-5 has many experimental facilities, including two graphite thermal columns, one of which is shown in Fig. 13.14. Immediately beneath the reactor tank, in the graphite zone, there is a space which can be utilized for isotope production or for other purposes as required.

Ordinary-Water-moderated Reactors

Materials Testing Reactor (MTR) *

Fuel: 4 kg (9 lb) uranium-235 (as aluminum alloy)
Moderator: ordinary water
Reflector: beryllium (~1½ ft) + graphite (~3½ ft)
Shield: two 4-in. steel plates (thermal shield) + 9 ft barytes concrete
Over-all dimensions: 32½ × 34 × 32 ft high
Control: up to 8 shim-safety, 2 regulating rods
Coolant: circulating water (20,000 gal/min)
Power: 30,000 kw
Thermal flux (av): 2×10^{14} neutrons/(cm²)(sec)

13.59. As its name implies, the Materials Testing Reactor (MTR), at Arco, Idaho, was designed to provide a high neutron flux for experimental purposes, in particular for the study of the effects of radiation on various reactor materials (see Chapter VIII). Operation of the reactor was commenced in March, 1952, and its special facilities have been extensively used since that time. The maximum accessible thermal flux of 4.5×10^{14} neutrons/(cm²)(sec), in the reflector near the core, is considerably higher than that of any other experimental reactor for which information is available.

13.60. Each of the curved fuel-bearing plates of the MTR consists of a sandwich of a 0.02-in. thick layer of an alloy of 10 to 20 per cent by weight of highly enriched (~93 per cent) uranium in aluminum, clad on each side with aluminum of the same thickness. The plates are approximately 2.8 in. wide and 2 ft long, and 18 plates, separated by 0.12 in. from each other, are brazed into aluminum side members to form a fuel assembly roughly 3 × 3 in. across, somewhat re-

* J. R. Huffman, *Nucleonics*, **12**, No. 4, 21 (1954); Report ORNL CF-53-12-21; S. McLain, *Proc. Kjeller Conf. on Heavy Water Reactors*, 1953, Jener Publications No. 7, p. 99.

sembling that shown in Fig. 13.13 for the CP-5 reactor. The mass of uranium-235 in an assembly is approximately 200 g and there are some 20 assemblies in the loaded reactor, located within a rectangular core of roughly 15 × 27 in. cross section. The bottom of each fuel assembly fits into a socket in a grid base, while another grid fits over the top, thus holding the units in place. The primary reflector of beryllium metal, about 1½ ft thick and 3 ft high, surrounds the core. The beryllium has a number of vertical holes to permit the flow of water for cooling the reflector. The core-reflector assembly is mounted in a cylindrical tank containing the water which acts as moderator-coolant and also as reflector at the top and bottom.

13.61. The section of the reactor tank around the core region is made of aluminum and has an inner diameter of 4½ ft. Above and below there are

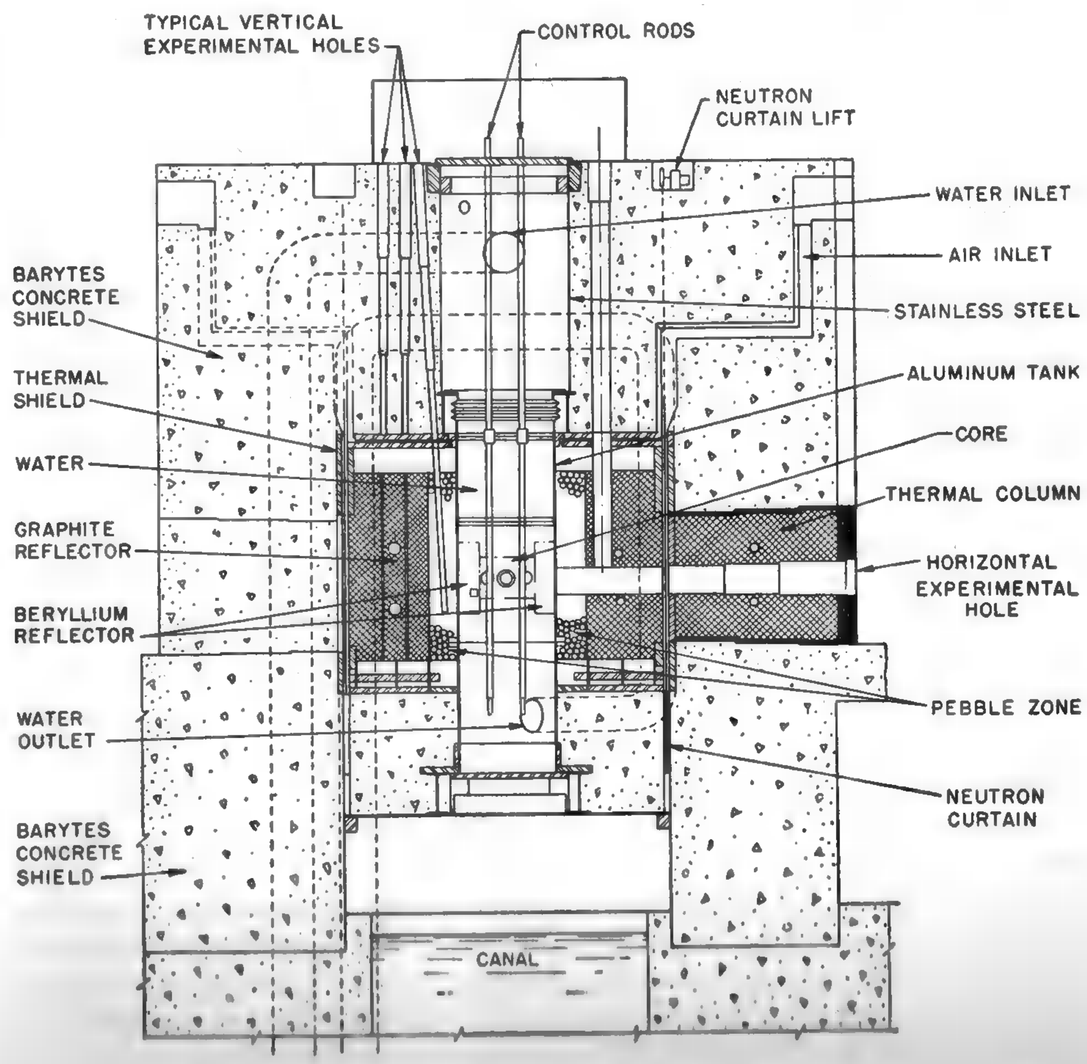


Fig. 13.15 Sectional drawing of the MTR at Arco, Idaho

stainless steel extensions, so that the whole forms a well, 30 ft deep, that is kept full of water during operation. For cooling purposes the water enters the well (at 100°F) about 15 ft above the core, flows between the fuel plates, and leaves (at 111°F) at some 5 ft below the core (Fig. 13.15).

13.62. To provide additional space for experiments, a secondary reflector of graphite is located outside the reactor tank. In view of the possibility of dimensional changes due to radiation, oxidation, and temperature, the graphite reflector is divided into two zones. There is, first, the pebble zone, $7\frac{1}{3}$ ft square and 9 ft high containing some 700,000 graphite spheres 1 in. in diameter. The pebbles can be removed, if necessary, through the discharge chute and then replaced without dismantling any part of the reactor. The second (or permanent) zone, made of graphite blocks, has external dimensions of 12×14 ft by $9\frac{1}{3}$ ft high. The graphite structure is penetrated by both vertical and horizontal holes for experimental purposes; some of the vertical holes are used for the passage of air which cools the reflector.

13.63. Close to the secondary reflector is the thermal shield, consisting of two 4 in. layers of steel; these are spaced 4 in. apart and cooling air flows through the gap. The 9-ft thick biological shield of barytes concrete (about 93 per cent barium sulfate in aggregate) surrounds the reactor at the sides. Above and below the core region the water serves as a shield, but in addition there are steel plugs containing 11 in. layers of lead shot, at both top and bottom of the reactor tank. The driving mechanisms for the control rods are mounted in the top plug.

13.64. The shim-safety control rods, which may be up to eight in number, move vertically in spaces within the fuel lattice. Each of these rods has an upper section containing cadmium and a lower one consisting of fuel plates; this type of design makes a valuable contribution to the neutron economy, as explained in § 6.7. The shim-safety rods are held up by electromagnets so that they will fall under the influence of gravity (and the pressure of the cooling water flow) when the exciting current is interrupted by the appropriate signal or if there is a power failure (cf. Table 6.4).

13.65. Two vertical regulating rods of cadmium, sandwiched between steel tubing, are located in the beryllium reflector close to the reactor core. Only one rod is operated at a time, the other being a spare. The motion of these rods can be controlled manually or automatically to keep the reactor power at any desired level. The servo-system described in § 6.125, *et seq.*, for bringing a reactor up to its operating power and keeping it at that level, is essentially that used in the MTR.

13.66. In accordance with its primary function as a research tool, there are over 100 experimental facilities in the MTR (cf. Fig. 13.16). Some of these extend into (or through) the primary beryllium reflector, whereas others penetrate only into the graphite zone. One of the narrower faces of the reactor is penetrated by a thermal column, and it was originally planned to have another one on the opposite face. Subsequently, it was decided to assign the space to

shielding studies, but it has not been used for this purpose and it is at present filled with blocks of barytes concrete.

13.67. Loading and unloading the reactor with fuel is performed with manually operated grappling tools through 20 ft of water. The spent fuel assembly is lifted just above the beryllium reflector and moved horizontally to a position directly over a hole in the reflector that is normally filled with a beryllium plug.

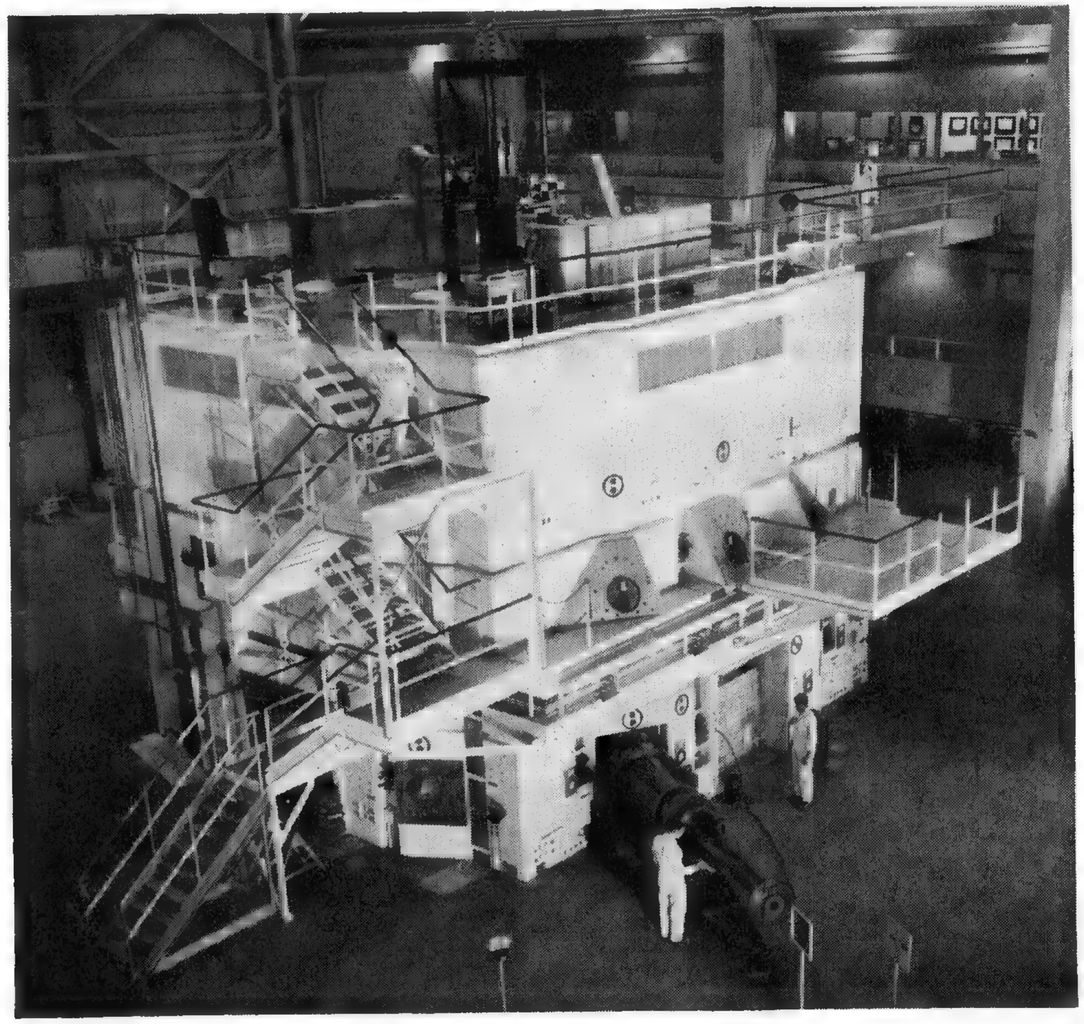


FIG. 13.16 General view of the MTR

The fuel assembly is then lowered into a water-filled discharge tube from which it is transferred by a special unloading device into a canal containing an 18-ft depth of water. In reloading the reactor the new fuel assemblies are lowered into place by hand from the top of the reactor.

13.68. One of the problems arising in the operation of the MTR is the increase in xenon poisoning after shut-down; as seen in Chapter V, this may become

very considerable in a reactor having a thermal flux as high as 2×10^{14} neutrons/(cm²)(sec). Some excess reactivity allowance for xenon override is made in the fuel loading (cf. Table 6.1, last column), but normally this is sufficient only for the first 30 min (or so) after shutdown. If the MTR cannot be restarted within this period, there may be a delay of about 2 days before it can again become operative. However, it is always possible to restart the reactor at any time by loading fresh fuel elements into the core.

Low-Intensity Test Reactor (LITR)

13.69. The mock-up of the MTR at the Oak Ridge National Laboratory, which was used first for mechanical and hydraulic studies, and later for criticality measurements, was subsequently converted into a general experimental facility and named the Low-Intensity Training Reactor (LITR).* It is essentially a low-power model of the MTR and normally operates at 3000 kw. The average thermal flux is about 2×10^{13} neutrons/(cm²)(sec). The new Omega West Reactor at Los Alamos Scientific Laboratory is to be of the LITR type; it is expected to operate at a power of 1000 to 4000 kw.

Swimming Pool Type Reactor†

Fuel: 2.5 to 3.5 kg (5.5 to 7.7 lb) uranium-235 (in aluminum alloy)
Moderator: water
Reflector: water (+ beryllium, beryllium oxide, or graphite in some cases)
Shield: water + concrete tank
Over-all dimensions: $20 \times 40 \times 22$ ft high (variable)
Control: 2 shim-safety, 1 regulating
Coolant: water (free-convective flow up to 100 kw)
Power: 100 kw (or more)
Thermal flux (av): 5×10^{11} neutrons/(cm²)(sec)

13.70. The prototype of the so-called Swimming Pool Reactor (also referred to as the Low Cost Reactor) is the Bulk Shielding Reactor (BSR) completed at Oak Ridge National Laboratory in December, 1950. It is, to a certain extent, a highly simplified version of the MTR that is inexpensive to construct and easy to operate. Because of these attributes, the Swimming Pool Reactor is regarded as one that is particularly suited to university and industrial-type research laboratories (§ 12.27). The construction of several such reactors is now being planned or is actually under way, and models are available commercially (Fig. 13.17).

13.71. The fuel elements are of the flat sandwich-plate type, consisting of an alloy of highly enriched uranium and aluminum clad on both sides with

* *Nucleonics*, **10**, No. 6, 75 (1952).

† Reports AECD-3435, 3557; W. M. Breazeale, *Nucleonics*, **10**, No. 11, 56 (1952); Report ORNL-991; *Chem. Eng. Prog. Symposium Series*, No. 12, **50**, 6 (1954); see also L. C. Widdoes, *ibid.*, No. 11, **50**, 195 (1954); E. H. Krause, *ibid.*, No. 11, **50**, 203 (1954); J. J. O'Connor and L. S. Foster, *ibid.*, No. 12, **50**, 59 (1954); H. J. Gomberg, Report AECU-2000, p. 51.

aluminum. A fuel assembly is approximately 3 in. square and 3 ft long, and is made up of from 10 to 18 fuel plates, of about 0.06 to 0.10 in. total thickness, spaced equally. The length of the fuel-bearing region is close to 2 ft. Normally 16 to 25 fuel assemblies are required, depending on the type and efficiency of the reflector. In many designs the reflector is removable, for flexibility in adjusting the lattice dimensions and in performing experiments. A loaded lattice, with beryllium oxide reflector contained in dummy fuel boxes, is shown in Fig. 13.18.* Above the lattice can be seen the three control-rod holders, in slotted cylindrical guides, and a number of ionization chambers for neutron flux measurement. The lattice and control system are suspended from a bridge across the top of the water tank ("Swimming Pool") in which the core is immersed.

13.72. The water in the concrete tank, which covers the reactor core to a depth of about 20 ft, serves as moderator, reflector, coolant, and as a shield. Additional shielding at the sides is provided by the concrete wall of the pool.

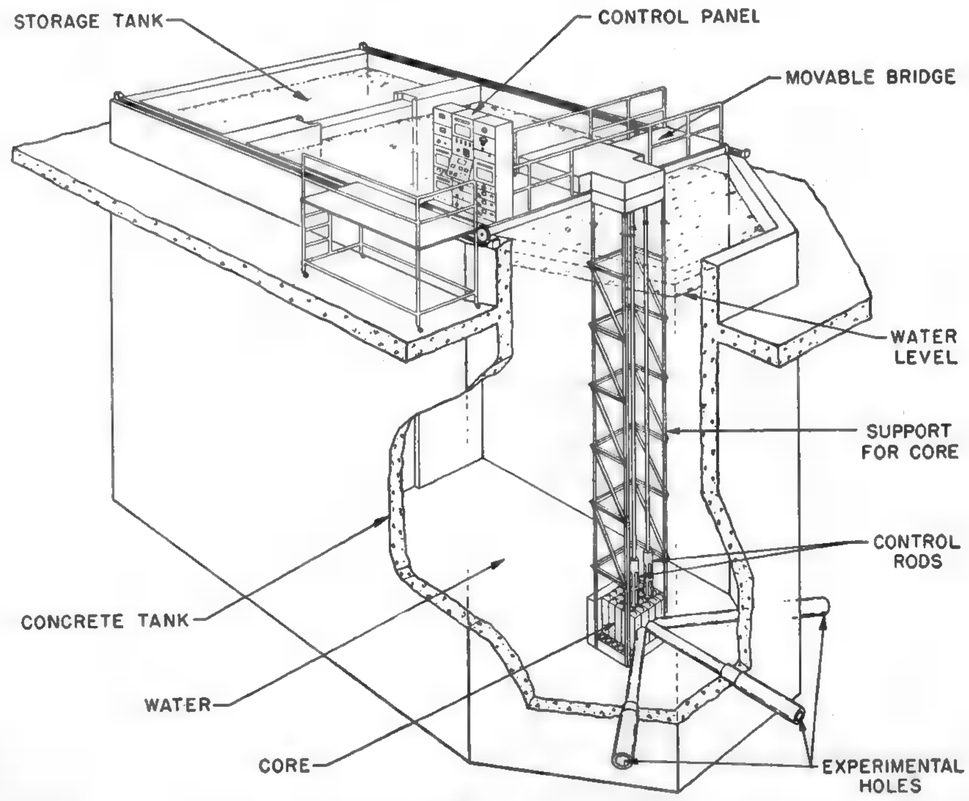


FIG. 13.17 Cutaway drawing of a Swimming Pool reactor. (Pennsylvania State University)

* Although beryllium oxide is a better reflector, graphite, which is less expensive, provides a good alternative.

In some versions of the Swimming Pool reactor the tank is built into the side of a hill, so that the earth contributes to the shielding.

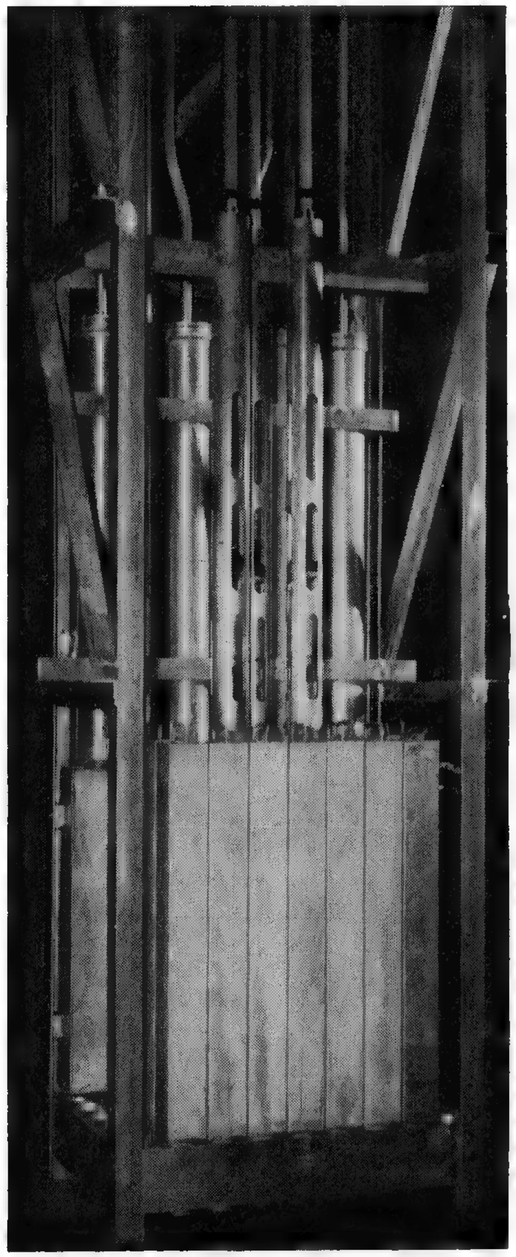


FIG. 13.18 Core of Swimming Pool reactor at Oak Ridge National Laboratory. (Oak Ridge National Laboratory)

13.73. The control rods, which may contain cadmium or boron (or boron carbide), move vertically in spaces within the core similar to those occupied by the fuel boxes. An iron armature or core piece is fastened to the top of each rod and the armature and rod are lifted by an electromagnet. When the exciting current to the magnet is reduced, the rods drop and shut the reactor down. The regulating rod can be adjusted so as to maintain the desired power level, either by manual operation or automatically by means of a servomechanism.

13.74. The design of the fuel elements permits satisfactory operation at 100 kw,* with a margin of safety, using free-convection cooling by the water, to remove the heat generated in the core. If, due to accidental circumstances, the power level rises above 1500 to 2000 kilowatts, the water between the fuel plates will boil. The steam produced will then displace sufficient moderator to inhibit runaway; the reactor is thus inherently safe, provided, as is invariably the case, it does not have too much available excess reactivity. Although power levels up to about 1000 kw are possible with natural-convection cooling, water circulation is recommended above 100 kw because of the radioactivity due to nitrogen-16. It is estimated that a flow rate of about 500 to 900 gal/min will permit safe operation up to 1000 kw with approximately 15°F rise in temperature of the water.

13.75. For experimental studies the apparatus or material can be lowered into the pool and placed in contact with the reactor face or in the core. Alternatively, beam holes may be provided through the walls of the tank (see Fig. 13.17). A thermal column can be added to the Swimming Pool reactor if required.

* It is estimated that at this power some forms of the Swimming Pool reactor could operate for something like 60,000 hr before the fuel elements would need to be replaced.

*Experimental Boiling Water Reactor**

13.76. The advantages of a boiling-water reactor for the direct production of steam were considered in § 12.149, *et seq.*, and it was mentioned that preliminary experiments have demonstrated the self-regulating property, up to certain power levels, of such reactors. After the destruction of the core of the first installation in July, 1954, a second experimental boiling-water reactor was constructed at Arco, Idaho to test the operational reliability of the boiling-water system at higher pressures. This reactor was completed in October, 1954; it operates at a (heat) power of 6000 kw and a pressure of 300 psi of steam.

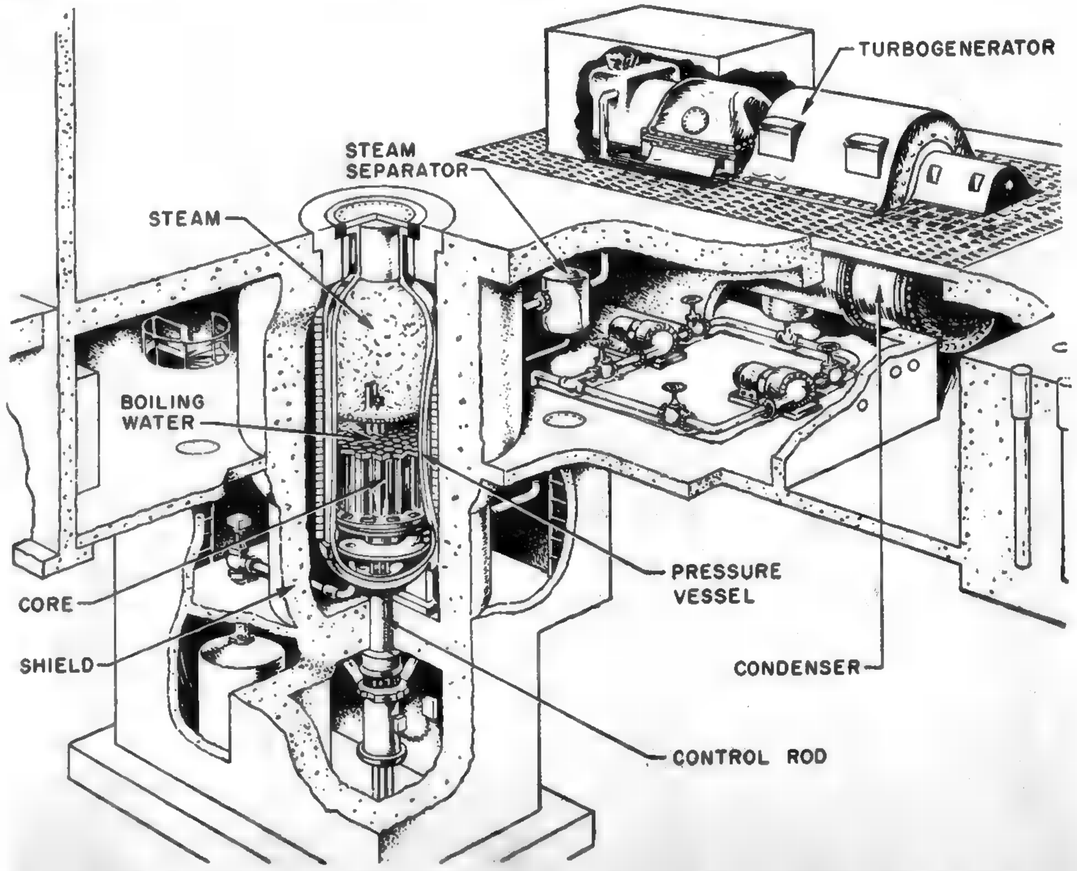


FIG. 13.19 Schematic diagram of power production from boiling-water reactor. (Argonne National Laboratory)

* See pp. 11 and 15 of Report referred to in § 13.28; S. Untermyer, *Nucleonics*, **12**, No. 7, 43 (1954); also, Seventeenth Semiannual Report of the Atomic Energy Commission, U. S. Government Printing Office, January, 1955, pp. 22–23.

13.77. Based on the successful functioning of the experimental installations, an Experimental Boiling-Water Reactor (EBWR) is being planned for construction at the Argonne National Laboratory. The reactor will use zirconium-clad, plate-type fuel elements; most of these will contain natural uranium with a few of enriched material, and ordinary water as the moderator-coolant. The steam produced will be fed directly to a turbogenerator in the conventional manner (Fig. 13.19). The heat power level of the reactor is to be about 20,000 kw and an electrical output of 5000 kw is to be expected, according to preliminary specifications. Among the objectives of the EBWR is the construction of a leakproof system, which is desirable in any event and would be particularly important if heavy water were used as moderator-coolant. It is also desired to determine whether significant amounts of radioactive material are deposited in the turbine, condenser, pumps, etc.

Pressurized-Water Power Reactors

13.78. The prototype (Mark I) of the Submarine Thermal Reactor (STR) was probably the first full-scale power reactor to be completed; it started operation at the National Reactor Testing Station, Arco, Idaho, in March, 1953 and produced electrical power in June of that year. Apart from the facts that it uses zirconium-clad, enriched uranium as fuel and pressurized water as the moderator-

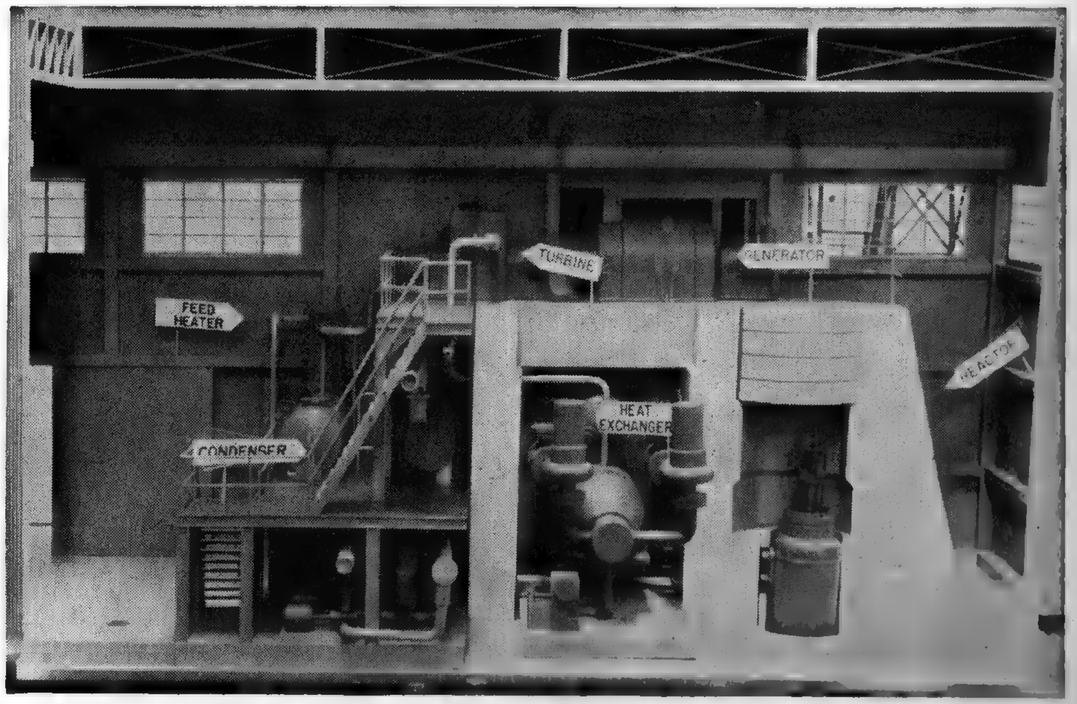


FIG. 13.20 Model of Army Package Power Reactor plant. (U. S. Army Signal Corps)

coolant, essentially no information about this reactor has been released.* However, the success of the STR as a means for propelling a submarine was fully demonstrated when the *U.S.S. Nautilus* commenced its sea trials on January 17, 1955.

13.79. Another power reactor which is to use pressurized water as moderator-coolant and, obviously, enriched uranium as fuel is the Army Package Power Reactor (APPR),† having transportable components, referred to in § 12.51. The maximum size of any fully fabricated part is to be $9 \times 9 \times 27$ ft and its weight is not to exceed 10 tons; the over-all size of the complete power unit is tentatively estimated as 80 ft long, 29 ft wide, and 42 ft high (Fig. 13.20). The reactor has been designed to produce 10,000 kw of heat, and the electrical output is expected to be at least 1700 kw. It is reported that the pressure of water in the reactor will be about 1200 psi; the water will enter at 432°F and leave at 450°F. The saturated steam from the heat exchanger will be at about 380°F and 200 psia. An important feature of the reactor is to be the long life of the fuel elements which will probably be clad with stainless steel. A contract has been let for the construction of a prototype of the APPR to be built at Fort Belvoir, Virginia; operation is expected by 1958.

13.80. The power reactor concerning which most data are available is the Pressurized Water-Reactor (PWR) being built near Shippingport, Pennsylvania; this is to be the first central-station nuclear power plant in the United States.‡ Pressurized water will be the moderator-coolant and the fuel is to be slightly enriched uranium containing 1.5 to 2 per cent of uranium-235. According to the tentative specifications, the core will consist of 15 to 20 tons of uranium, made up of closely spaced, zirconium-clad fuel elements, in the form of a right circular cylinder 6 ft diameter and $7\frac{1}{2}$ ft high. The pressure vessel holding the core is expected to be about 9 ft in diameter and 28 ft high.

13.81. The power output of the PWR will be at least 260,000 kw of heat. The maximum heat flux will be greater than 350,000 Btu/ft², the average power density will be 45 kw/liter, and the specific power 1000 kw/kg of fissionable material. The inherent dynamic stability of pressurized water reactors of proper design will be the basis of control during power changes. As a result of the large negative temperature coefficient of reactivity, the temperature of the water leaving the reactor during operation should be independent of small variations in the power demand (cf. § 6.153).

13.82. The pressure in the reactor vessel will be about 2000 psig, at which the saturation temperature of water is nearly 640°F; the fuel surface temperature

* *Nucleonics*, **10**, No. 8, 72 (1952); L. H. Roddis, Jr., and J. W. Simpson, *Westinghouse Engr.*, **15**, No. 2, 74 (1955).

† *Nucleonics*, **12**, No. 10, 22 (1954); see also R. S. Livingston and A. L. Boch, *ibid.*, **13**, No. 5, 24 (1955).

‡ See p. 10 of Report referred to in § 13.28; also, C. H. Weaver, Atomic Industrial Forum Report, July, 1954, p. 16.

should thus not exceed 600°F. The cooling water temperature will be in the range from 500° to 550°F and steam will be produced at 600 psig; at this pressure the saturation temperature is about 490°F.

13.83. Four independent heat-transfer loops will be available for steam production (Fig. 13.21), although only three will be used at a time with the fourth as standby. In each loop water will be circulated at the rate of 16,000 gal/min and the total pumping power required will be 3600 kw. The electric power generated should be at least 60,000 kw.

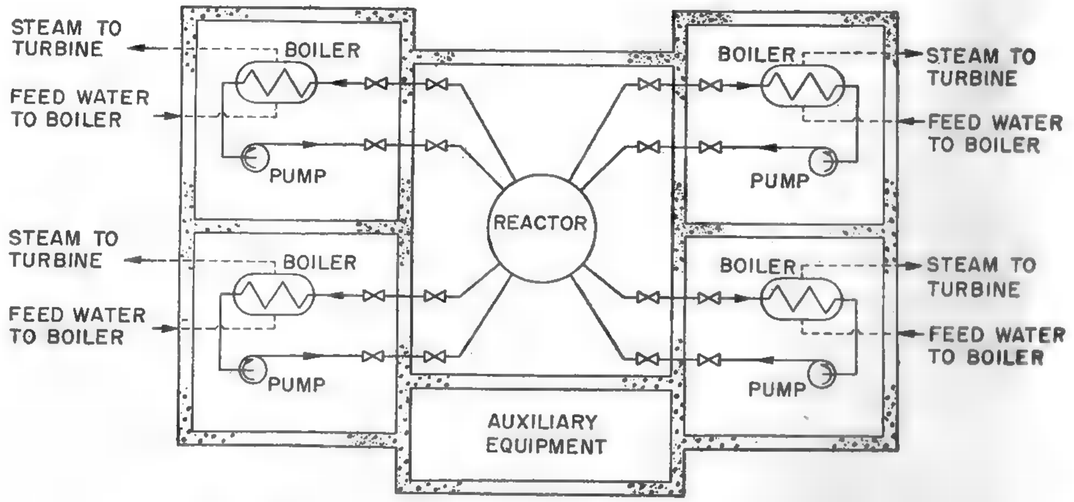


FIG. 13.21 Ground plan of Pressurized Water Reactor and Boilers at Shippingport, Pennsylvania. (Westinghouse Atomic Power Division)

13.84. As a precautionary measure, in the unlikely event of an accident, all parts of the system under pressure, i.e., the reactor vessel and the heat exchangers (steam generators), will be enclosed in strong gas-tight containers. These will all be located underground in concrete and steel structures, to provide shielding and to minimize the spread of radioactivity in the event of an accident (Fig. 13.22).

13.85. In addition to supplying valuable information concerning the economics of nuclear power, the PWR will contribute to the technology of pressurized water and will provide facilities for testing fuel elements capable of a high percentage burn-up. The project will also demonstrate whether large pressure vessels can be built to the specifications required for reactor operations, and methods of loading and unloading the fuel elements through the pressure shell will be developed. The results will have an important bearing on the design of power reactors of the future.

HOMOGENEOUS WATER-MODERATED REACTORS

*The Water Boiler Reactor**

Fuel: 0.87 kg (1.91 lb) uranium-235 in 12.7 liters solution
Moderator: water
Reflector: ~4½ ft graphite
Shield: ½ in. B_4C in paraffin, 2 in. steel, 4 in. lead, 5 ft concrete
Over-all dimensions: 15 × 15 × 11 ft high
Control: 3 shim-safety in reflector, 2 regulating in core
Coolant: water at ~60°F (3 gal/min)
Power: 45 kw
Thermal flux (av): 10^{12} neutrons/(cm²)(sec)

13.86. The Water Boiler Reactor at Los Alamos Scientific Laboratory was the first reactor to use water as a moderator and also the first reactor to have the

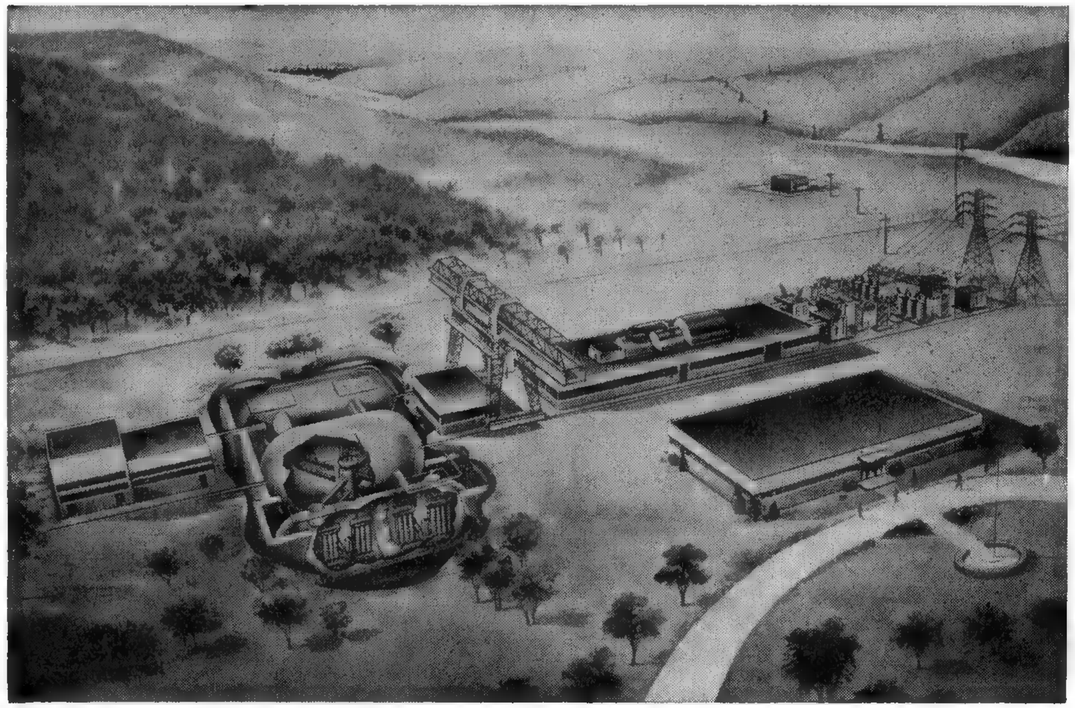


FIG. 13.22 Artist's sketch of over-all PWR plant, showing reactor and boilers in underground, gas-tight containers. (Westinghouse Atomic Power Division)

fuel in solution. The object of the original form of the reactor, known as LOPO (low power), was to provide experience in the operation and control of a chain reacting system using a minimum of enriched fuel. The reactor, which went critical in May, 1944, contained 565 g of uranium-235, in the form of uranyl

* L. D. P. King, *et al.*, Report AECD-3287; Report AECU-2000, p. 81; *Rev. Sci. Instr.*, **22**, 489 (1951); see also Reports AECD-3059, AECD-3063, AECD-3065.

sulfate enriched to the extent of 14.6 per cent in the fissionable isotope, dissolved in some 13 liters of water, contained in a stainless steel (Type 347) sphere of 1-ft diameter and $\frac{1}{32}$-in. wall thickness. The sphere was surrounded by beryllium oxide as reflector, since calculations indicated that the critical mass of uranium-235 would be minimized by using this in preference to graphite. Because of the lack of a shield and cooling system, the operating level of LOPO did not exceed the very low power of 50 milliwatts.

13.87. Following upon successful experience with LOPO, it was decided to modify the reactor, by providing forced cooling and a shield, to permit operation at a considerably higher power. Due to various causes, e.g., a thicker sphere ($\frac{1}{16}$ in.), internal cooling coils, experimental facilities, and the substitution of part of the beryllium oxide reflector by a graphite thermal column, the critical mass of the uranium-235 (14.0 per cent enrichment) was increased to 808 g in about the same volume (13.65 liters) of solution.* Another factor contributing to the increase in the critical mass was the use of uranyl nitrate in place of the sulfate, since nitrogen has a higher neutron absorption cross section than sulfur. The change was made from sulfate to nitrate in case it became necessary to recover the fissionable material at short notice; it was felt, at the time, that the nitrate could be more readily extracted with ether.

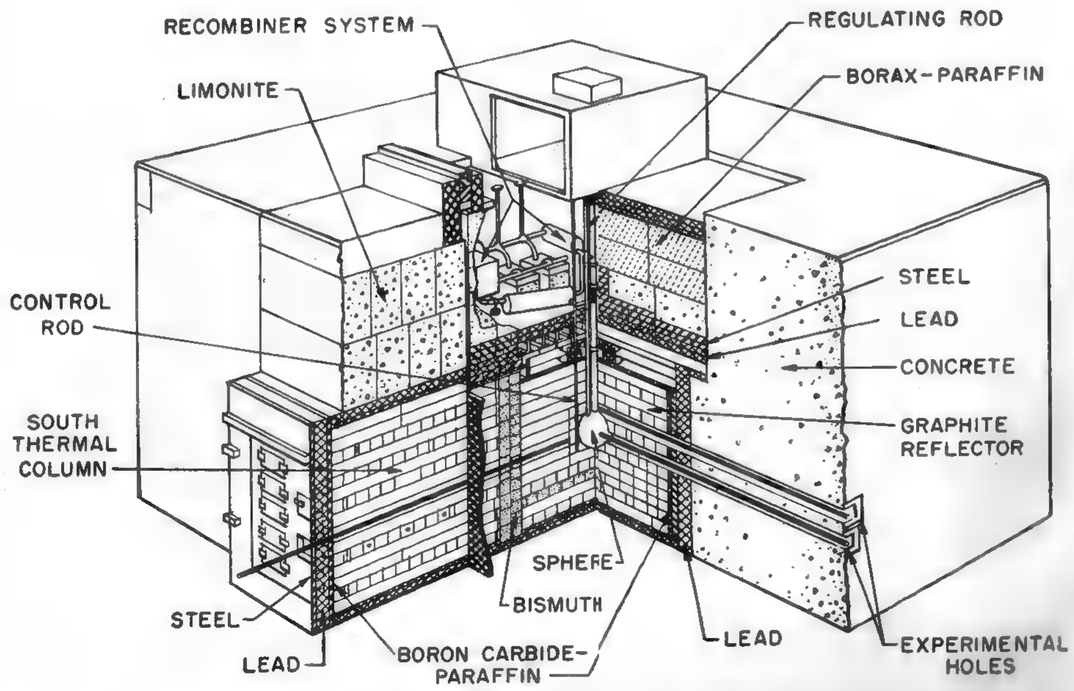


FIG. 13.23 Cutaway drawing of Water Boiler Reactor at Los Alamos Scientific Laboratory. (Los Alamos Sciertific Laboratory)

* In October, 1946, the amount of uranium-235 was increased further to 870 g (14.5 per cent enrichment).

13.88. The modification of the Water Boiler, called HYPO (high power), commenced operation in December, 1944. Its normal power was 6 kw and the average thermal flux about 10^{11} neutrons/(cm²)(sec). The temperature of the solution during operation was about 175°F with the cooling water (at 46°F) flowing at the rate of about 50 gal/hr.

13.89. In 1949 a series of changes were commenced with the object of increasing the experimental facilities, the safety, and operating power of the Water Boiler. These modifications were completed in March, 1951, and a drawing of the final form, known as SUPO (super power), is shown in Fig. 13.23. The data given at the head of this section refer to this particular model of the Water Boiler reactor.

13.90. The chief design changes in SUPO were the following: (1) The beryllium oxide reflector was replaced by graphite; this permits a more rapid shut-down of the reactor and eliminates the variable starting source of neutrons due to the (γ, n) reaction on beryllium (cf. § 2.74). (2) The enrichment of the uranium-235, as uranyl nitrate, was increased to 88.7 per cent (instead of the former 14.5 per cent). This made possible a decrease in the total nitrate concentration and so reduced the formation of nitric acid and the consequent gas evolution due to

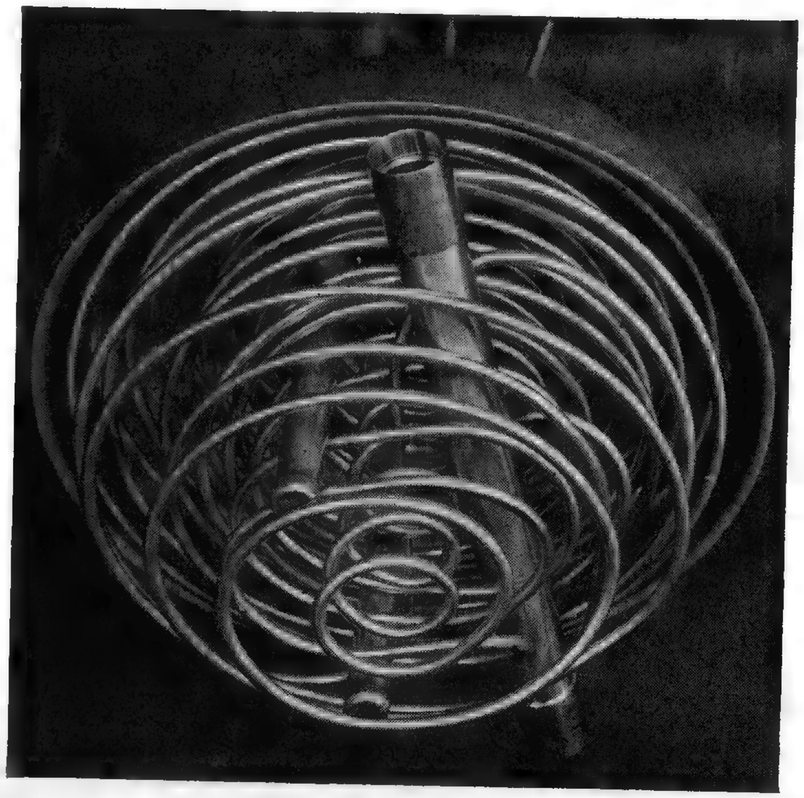


FIG. 13.24 Half of sphere of Los Alamos Water Boiler showing cooling coils and "glory hole" tube. (Los Alamos Scientific Laboratory)

radiation decomposition. Further, the increase in enrichment of uranium made it possible to operate the reactor without appreciably more uranium-235, in spite of the poorer reflector and other changes which are disadvantageous to the neutron economy. (3) The former single cooling coil was replaced by three 20-ft long, $\frac{1}{4}$-in. O.D., $\frac{3}{16}$-in. I.D. stainless steel tubes (Fig. 13.24), thus permitting an increase in the operating power of the reactor.

13.91. In the HYPO form of the Water Boiler, the cadmium control rods were located in the beryllium oxide reflector. With the change to graphite, a less effective reflector, it was decided to have two other rods which could be inserted into the core. These rods contain boron, enriched in boron-10, and enter the stainless steel sphere containing the uranium solution through reentrant thimbles. In addition to these two rods, which are used for regulating purposes, there are three cadmium shim-safety rods in the graphite reflector.

13.92. Because of its large negative temperature coefficient, the Water Boiler is inherently self-regulating. Nevertheless, continuous but transient variations in power occur, e.g., due to gas bubbles and convection currents in the core solution. Since steady neutron fluxes are essential for many experiments, automatic control of the regulating rods is used to maintain the power level constant within 0.1 per cent.

13.93. An important feature of SUPO is the recombination system for handling the explosive hydrogen-oxygen mixture resulting from decomposition of the water by radiation. A stream of circulating air (3.5 ft³/min) carries the gases up the vertical stack attached to the top of the reactor sphere, then through the recombiner, as described in § 8.149; the water formed is returned to the reactor so that there is very little loss. The use of this system not only removes the hazard of an explosion, but it also minimizes changes in the concentration of the fuel solution. There is, however, some loss due to the escape of oxides of nitrogen, and this is made up by addition of small amounts of nitric acid from time to time.

13.94. Among the experimental facilities is a 1-in. (internal) diameter tube, referred to as a "glory hole," running right through the reactor sphere; this is seen in the photograph in Fig. 13.24. In addition, there is a $1\frac{7}{16}$-in. (internal) diameter tube which runs completely through the reactor tangential to the sphere. The Los Alamos Water Boiler has two graphite thermal columns, on the north and south faces, respectively (see Fig. 13.23). In front of each of these is a "neutron window," consisting of an $8\frac{1}{2}$ in. layer of bismuth; this absorbs gamma rays, while permitting neutrons to pass through.

13.95. As may be seen in Fig. 13.23, the shielding varies somewhat at different locations. However, basically, it consists of $\frac{1}{2}$ in. of boron carbide in paraffin, followed by 2 in. of steel, 4 in. of lead, and 5 ft. of concrete. The thermal columns, which extend through the concrete, are shielded with boron carbide, lead, and steel. Several portions of the shield are removable to permit insertion of experimental equipment.

Other Water Boilers

13.96. The simplicity of construction of the Water Boiler reactor has made it of interest as a laboratory tool for experimental work with neutrons, and also to provide some training in reactor operation. A reactor of this type has been in operation at North Carolina State College, Raleigh, North Carolina, since 1953.* It differs from the Los Alamos Water Boiler in using uranyl sulfate (93 per cent enriched), instead of the nitrate, as the fuel material. Further, the vessel holding the solution is a cylinder rather than a sphere. Four stainless steel coils, each 7 ft long and $\frac{1}{4}$ in. inside diameter, carry cold (68°F) water through the core at the total rate of 4 gal/min, for cooling the fuel solution. The maximum temperature of this solution, at the operating level of 10 kw, is about 176°F. The average thermal flux is 2×10^{11} neutrons/(cm²)(sec).

13.97. Several versions of the Water Boiler reactor are being offered commercially. One of these, called the Water Boiler Neutron Source (WBNS), was designed primarily, as its name implies, as a laboratory source of neutrons.† It has a low power and is compact, simple, safe, and relatively inexpensive. The device operates at powers up to 1 or 2 watts and the average thermal flux is then about 2×10^7 neutrons/(cm²)(sec).

The Homogeneous Reactor Experiment ‡

13.98. Because of the attractive features of homogeneous reactor systems for power production (§ 12.132), the Homogeneous Reactor Experiment (HRE) was designed to test the feasibility of maintaining a fission chain reaction in an aqueous circulating solution at high temperatures and power levels. The pilot reactor at Oak Ridge National Laboratory attained criticality in April, 1952, and was operated at powers up to 1000 kw. Through its associated turbogenerator system, it first produced electricity in February, 1953, this being the second known instance of actual electric power production from nuclear energy.§ During 1954, the HRE, having served its purpose, was dismantled to make room for HRE-2 or Homogeneous Reactor Test (HRT), a homogeneous reactor experiment on a larger scale (§ 13.105). Because of its historical significance in the development of nuclear power, the original HRE reactor will be described as if it were still in existence.

13.99. The core of the HRE is a Type 347 stainless steel sphere, 18 in. in diameter, through which an aqueous solution of a 93 per cent enriched uranyl sulfate is circulated at the rate of 100 gal/min. At the maximum operating level of 1000 kw, the fuel solution leaves the core vessel at 482°F, passes through

* C. K. Beck, *et al.*, Report ORO-33, Report AECU-1986; *Nucleonics*, **7**, No. 6, 5 (1950).

† H. Jarvis and R. Chalker, *Proc. Conf. on Nucl. Eng.*, Berkeley, 1953, B-1; M. E. Remley, *ibid.*, B-13; *Science*, **119**, 9, 21 (1954); H. Pearlman, Report AECU-2900, p. 183; see also, *Chem. Eng. News*, **30**, 3506, 3642 (1952).

‡ S. E. Beall and C. E. Winters, *Chem. Eng. Prog.*, **50**, 256 (1954); A. M. Weinberg, Atomic Industrial Forum Report, July 1954, p. 26.

§ The first known instance was the EBR (see § 13.122).

a steam generator (boiler), and is returned at 410°F. A total pressure of 1000 psi is maintained in the fuel system by heating a small volume to 545°F in a pressurized chamber directly above the core (Fig. 13.25). Although the pressure is considerably more than is required to prevent boiling of the fuel solution, it is necessary in order to minimize the volume of the gases due to radiation decomposition.

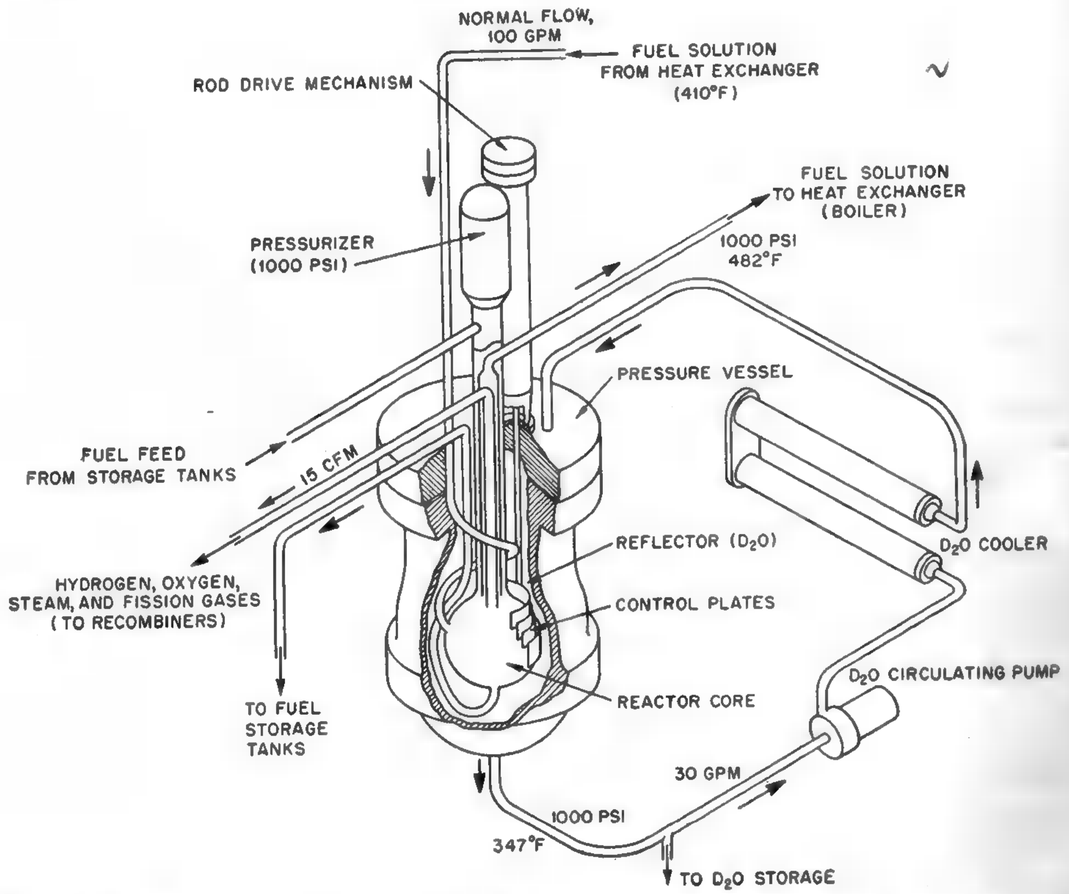


FIG. 13.25 Schematic representation of Homogeneous Reactor Experiment at Oak Ridge National Laboratory. (Oak Ridge National Laboratory)

13.100. In the HRE the reflector is a 10-in. layer of heavy water surrounding the core vessel. A pressure of helium, within 100 psi of that of the fuel solution, is maintained over the reflector to decrease stresses in the $\frac{3}{16}$ in. thick wall of the fuel container. Both fuel and reflector are contained within an outer pressure vessel of 39 in. internal diameter and 3 in. wall thickness. The heat in the reflector (about 5 per cent of the total), due mainly to transfer from the core, is removed by circulating the heavy water through a heat exchanger which serves as the boiler feed-water heater. In this manner, the temperature of the reflector is maintained at about 350°F.

13.101. The hydrogen-oxygen gas mixture formed by radiation decomposition, at the rate of 10 ft³/min (at S.T.P.) at a power of 1000 kw, is continuously separated from the fuel solution by vortex action in the core. After cooling and separating from entrained liquid, the gases pass to a flame recombiner and catalytic bed, as described in § 8.150. In the HRE, no attempt was made to utilize the 40 kw of heat liberated in the recombiner. It has been suggested that this high-temperature heat might be used to superheat steam, but the amount is probably not large enough to have any significant effect on the economics of power production.

13.102. Control of the HRE is achieved in a number of unusual ways. The reactor is started up, for example, by increasing the concentration of the uranium salt in the fuel solution, and it is shut down by dilution, using the water condensate from the flame recombiner for the purpose. Changes in the power level are realized by altering the demand, e.g., on the turbogenerator driven by the 200-psi steam produced in the boiler. Upon increase of the generator load, there is an increase in the steam demand; there is, consequently, a decrease in the steam pressure and temperature. This is reflected in the return of the fuel solution to the reactor at a lower temperature. Because of the large negative temperature coefficient of the system, the reactivity increases very rapidly and the power rises, to compensate for the added load, until the original temperature is restored.*

13.103. A special feature of the HRE is that the operating temperature can be altered by changing the concentration of the fuel solution, rather than by means of control rods, as in solid-fuel reactors. At a concentration of about 25 g uranium-235 in 1 kg of water, for example, the temperature cannot rise above 77°F; if it did, the system would become subcritical because of the negative temperature coefficient of reactivity, and the chain reaction would die out. If the concentration is increased to 41 g of uranium-235 to 1 kg of water, the temperature can be raised to 480°F and the fission chain will still be maintained. Change in temperature can also be achieved in the conventional manner by use of mechanically operated controls if desired, but this appears to be unnecessary.

13.104. When the HRE was designed, various safety measures were incorporated, including neutron-absorbing control plates in the reflector. The remarkable dynamic stability of the system indicates that mechanical controls are probably not required in a reactor of the HRE type. Its inherent safety has been confirmed by experiments in which the reactivity has been suddenly increased in various ways. The accompanying rapid release of energy causes the temperature to rise, but the resulting decrease in reactivity stabilizes the system at a power level determined by the demand.

* It should be noted that the ability to control a reactor automatically by the external power demand is, in principle, a general characteristic of reactors having appreciable negative temperature coefficients (see § 6.143, *et seq.*).

13.105. The HRE has proved so successful that it is being replaced by HRE-2 (or HRT) to operate at a maximum power of 5000 kw.* One of the objectives of this experiment will be to obtain further information on the control of homogeneous reactors (see also § 12.133). The next step in the development of homogeneous reactors is to be one using uranium-233 as fuel with a thorium breeding blanket. The power output of this Homogeneous Thorium Reactor (HTR), according to preliminary specifications, is to be about 65,000 kw of heat, yielding some 16,000 kw of electricity. The core of the reactor will not be as large as that of a full-scale plant, but the thickness and concentration of the thorium blanket are to be the same as for a central-station thermal power-breeder of this type. Both the HRE-2 (or HRT) and the HTR are to have associated chemical equipment for processing the fuel solutions.

Reactor with Unusual Moderator

Thermal Test Reactor

13.106. An unusual type of experimental reactor is the low-power Thermal Test Reactor (TTR) at Knolls Atomic Power Laboratory.† It uses highly enriched (90 per cent uranium-235) fuel, and a hydrocarbon oil, polyethylene, water, and graphite serve as moderators. One of the main purposes of the TTR is the determination of neutron absorption cross sections by the "danger coefficient" method; this is based on measurements of the change in reactivity of a reactor resulting from the addition to the core of a known amount of the absorber.‡ The TTR is also a good source of thermal neutrons and is said to be relatively inexpensive and safe to operate. It requires 2.7 kg of uranium-235 to attain criticality.

13.107. The active region of the reactor is a 3-in. annular space between a cylinder of graphite (12 in. diameter), which serves as an internal thermal column (and reflector), and an outer cylinder of aluminum (18 in. diameter). This space contains twenty fuel-slug tubes, each 2 in. diameter and 2 ft long (Fig. 13.26). The interstices between the slug tubes and the container are filled with water. Each fuel slug consists of a number of enriched uranium-aluminum alloy disks, 0.040 in. thick and having a $\frac{3}{16}$-in. hole in the center, alternating with 0.20-in. polyethylene spacers, fitted on to an aluminum rod about 18 in. long. The slugs are immersed in a light paraffin-base oil in the slug tubes. The reactor core is surrounded by a graphite reflector to form (approximately) a cube of 5-ft edge.

13.108. The TTR is located in a cell of 10 × 15 ft floor area; this is surrounded by a 6-ft concrete wall on four sides. The ceiling is a 1-ft concrete slab. There is an opening, with a movable shield, in one wall of the cell for access to the

* See p. 13 of Report on A.E.C.'s Five-Year Power Reactor Program, referred to in § 13.28.
† Report AECD-3530; H. B. Stewart, *et al.*, *Nucleonics*, **11**, No. 5, 38 (1953).
‡ Cf. H. L. Anderson, *et al.*, *Phys. Rev.*, **72**, 16 (1947).

reactor when it is shut down. In another wall there is a 4 × 4 ft hole for the external thermal column.

13.109. Eight cadmium control rods, two for regulating and six for safety action, move in $\frac{1}{2}$-in. diameter guide-tubes located in the space between the fuel-slug tubes and the outer aluminum cylinder containing the reactor. The safety rods have a magnetic release mechanism such as is used in many other reactors. Shim control is achieved by means of six thin cadmium sheets, clad with iron, which are also in the space between the slug tubes and the outer container wall.

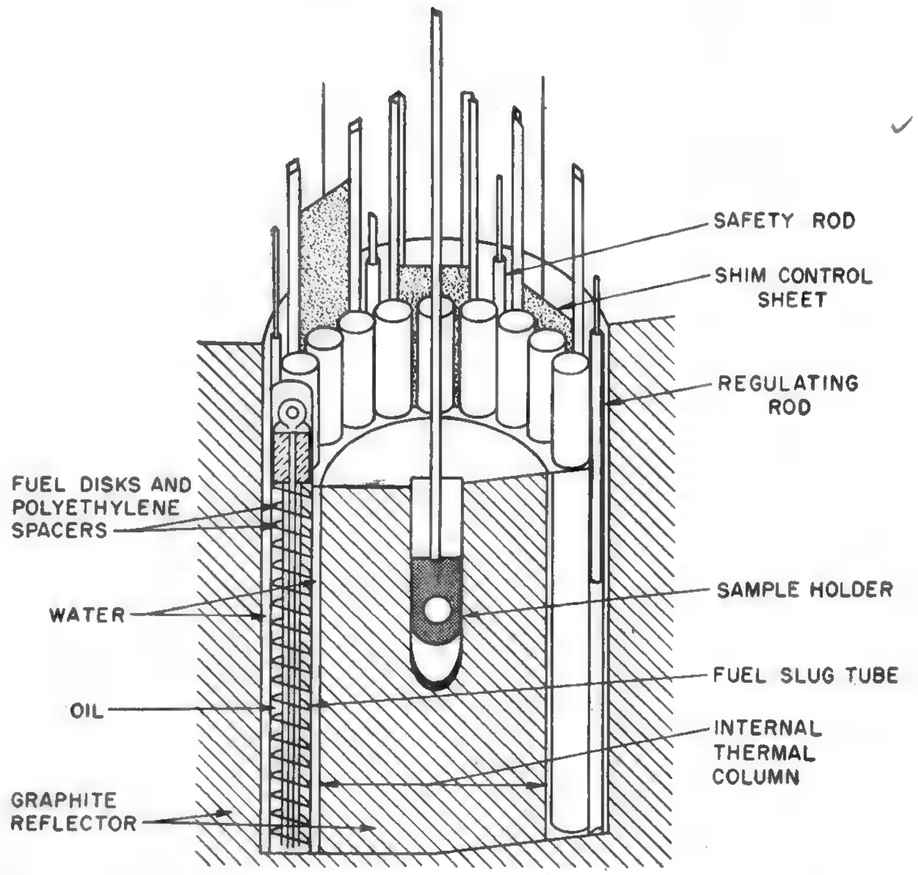


Fig. 13.26 Cross section of Thermal Test Reactor at Knolls Atomic Power Laboratory. (Knolls Atomic Power Laboratory)

The TTR has no forced cooling and the normal operating level is about 0.1 kw; the maximum flux, which occurs at the center of the internal thermal column, is 3.4×10^9 neutrons/(cm²)(sec).

13.110. The internal thermal column is one of the special features of the TTR. A slot in the center permits the insertion of experimental samples, for danger coefficient measurements, in the region where the thermal flux is highest. The change in reactivity of the reactor is then determined from the displacement of a

calibrated control rod. For general neutron studies use is made of the external thermal column which is an extension of the graphite reflector.

Fast Reactors

*The Los Alamos Fast Reactor**

Core: plutonium rods in 6 in. × 6 in. cylindrical cage
Moderator: none
Reflector: natural uranium and steel
Shield: Masonite, iron, boron plastic layers (30 in.) + 18 in. barytes concrete
Over-all dimensions: 11 × 15 × 9 ft high
Controls: 2 safety, 2 regulating in uranium reflector
Coolant: mercury
Power: 25 kw
Fast flux (max): 10^{13} neutrons/(cm²)(sec), av. energy 0.5 Mev

13.111. The first-known fast-neutron reactor was constructed at the Los Alamos Scientific Laboratory for experimental work requiring a good supply of fast neutrons.† It commenced operation in November, 1946, at a power of about 10 kw, but this was increased later (in March, 1949) to 25 kw. The reactor was dismantled during 1953 partly because of the failure of at least one of the fuel elements and the resulting contamination of the mercury coolant.

13.112. The reactor core consisted of a number of plutonium fuel rods, clad with steel, fixed vertically in a cylindrical steel frame, approximately 6 in. in diameter and 6 in. high. Originally, spacer rods of natural uranium were located between the fuel rods, but after the failure of one of these uranium rods in March, 1950, they were removed and replaced by dummy steel cans. The part of the reflector immediately surrounding the core consisted of blocks of natural uranium forming a cube of about 17-in. edge, with the reactor core in the center. Outside the uranium was a $\frac{1}{4}$-in. thick aluminum jacket containing tubes for cooling water, to remove the heat (about 1 kw) released in the uranium, and finally a 6-in. layer of steel to complete the reflector.

13.113. Around the reflector was a 4-in. layer of lead, and the whole, constituting a cube of about 38-in. side, was enclosed in a gas-tight, aluminum jacket, $\frac{1}{32}$ in. thick, to prevent escape of fission gases. A constant flow of helium through the aluminum-sealed system served to dilute and carry off any such gases that were released from the fuel or reflector.

13.114. The major portion of the shield, except in front of the thermal column, was made up of 3 in. of iron, 3 in. of pressed-wood fiber, 3 in. of iron, 3 in. of [illegible] plastic material containing boron, and then alternating 3-in. layers of iron and

* E. T. Jurney, *Nucleonics*, **12**, No. 9, 28 (1954); *Chem. Eng. Prog. Symposium Series*, No. 13, 50, 191 (1954).

† The reactor was nicknamed "Clementine," because of the association of "49," the code name for the fuel (plutonium), with the location of the reactor in a canyon near Los Alamos, New Mexico.

boron plastic to a total thickness of 30 in. Finally, there was an 18-in. thick wall of barytes concrete. There were some minor differences in the nature of the shield, e.g., at the top of the reactor, as may be seen in Fig. 13.27. Here, the shield consisted of removable layers of iron and boron plastic, and concrete blocks.

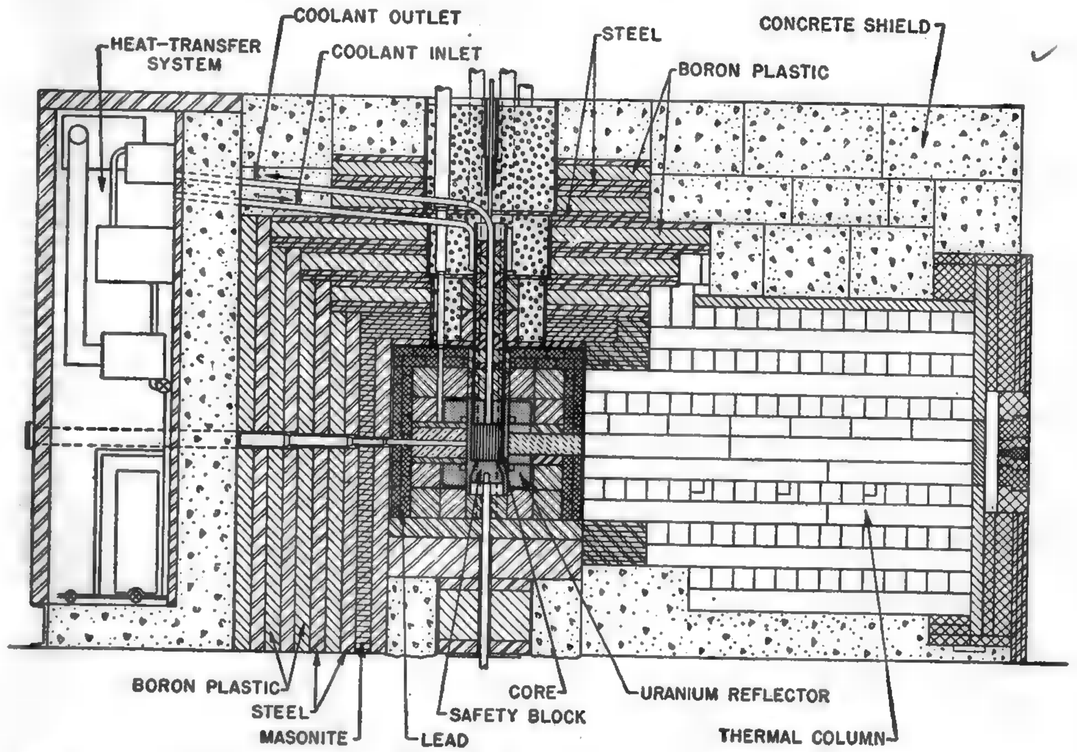


Fig. 13.27 Sectional drawing of Fast Reactor at Los Alamos Scientific Laboratory. (Los Alamos Scientific Laboratory)

13.115. Control was achieved by means of four cylindrical rods, consisting of a lower section of natural uranium and an upper section containing boron-10. These rods, each of which had a reactivity equivalent of 0.004, moved in vertical holes in the uranium reflector near the core. Two of the rods were for safety purposes, which would drop upon receipt of appropriate signals; as a result, part of the uranium in the reflector would be replaced by boron. The resulting loss of neutrons was sufficient to shut the reactor down. The other two rods were used for regulating the power level of the reactor. As an added safety measure, a 70-lb block of uranium, part of the reflector immediately below the core, was arranged so that it could drop 3 in. in an emergency.

13.116. In spite of the smaller fraction of delayed fission neutrons from plutonium-239 than from uranium-235 (see Table 2.11), so that the prompt-critical condition was attained more readily, control of the fast reactor did not prove difficult. The fact that the core had a negative temperature coefficient of reactivity (-1.5×10^{-5} per °F) made the reactor inherently stable.

13.117. Mercury was used as coolant because of its excellent properties as a heat-transfer agent; although the nuclei of this element readily absorb slow neutrons, the capture cross section for fast neutrons is very small. The mercury flowed through a steel pot, about 45 in. tall, at the bottom of which was the cage containing the fuel rods. It was not possible to drain the coolant from the core, otherwise the temperature might have become so high as to melt the plutonium. After leaving the pot at about 175°F, the mercury was circulated, by means of an electromagnetic pump, through a heat exchanger where it was cooled with water.

13.118. In addition to a thermal column, with its neutron window of bismuth, which made possible studies with slow neutrons, the reactor had a number of experimental holes for exposing materials to fast neutrons near the core. Well-collimated beams, having a fast-neutron flux of 10^9 neutrons/(cm²)(sec), could also be obtained outside the reactor. Near the hydrogenous portion of the shield, neutrons of intermediate energy were available.

*The Experimental Breeder Reactor**

Fuel: uranium-235 (core ~0.14 ft³ total volume)
Moderator: none
Reflector and breeding blanket: natural uranium
Coolant: NaK alloy (two circuits)
Power: 1000–1400 kw
Fast flux (av): 2×10^{14} neutrons/(cm²)(sec)

13.119. The Experimental Breeder Reactor (EBR), at Arco, Idaho, which commenced operation in 1951 (Fig. 13.28), was designed by the Argonne National Laboratory to obtain information concerning the possibilities of breeding with fast neutrons, with simultaneous power production using a liquid-metal coolant. Strictly speaking, the EBR is not a true breeder, since the fuel is uranium-235 and the fissionable product is plutonium-239.† However, it was evident that if a conversion ratio greater than unity could be achieved in this system, a true breeder, using plutonium as fuel, should be practicable.

13.120. The core of the EBR has been described as being "about the size of a regulation football"; this includes the almost pure uranium-235 fuel rods, as well as structural material and coolant. For the latter, sodium-potassium alloy (NaK) was chosen because of its satisfactory thermal properties, its temperature stability, and its low melting point, as a result of which it remains liquid at ordinary temperatures. Surrounding the core is a layer of natural uranium metal to act as reflector and breeding blanket; the uranium-238 captures fast neutrons escaping from the core with the ultimate formation of plutonium-239.

13.121. The coolant flows by gravity, at a rate of 300 gal/min, from a feed tank through the reactor, to a heat exchanger and then to the receiver tank (see Fig. 13.29). From the latter, the NaK is pumped back to the gravity feed tank. Since the sodium, in particular, becomes radioactive in its passage through the reactor, the whole of this portion of the cooling system must be shielded.

* W. H. Zinn, *Nucleonics*, 10, No. 9 8 (1952); Atomic Industrial Forum Report, July, 1954, p. 39; H. V. Lichtenberger, *Chem. Eng. Prog. Symposium Series*, No. 13, 50, 139 (1954).

† A core containing plutonium-239 as fuel was scheduled to be loaded into the reactor during the early part of 1955.

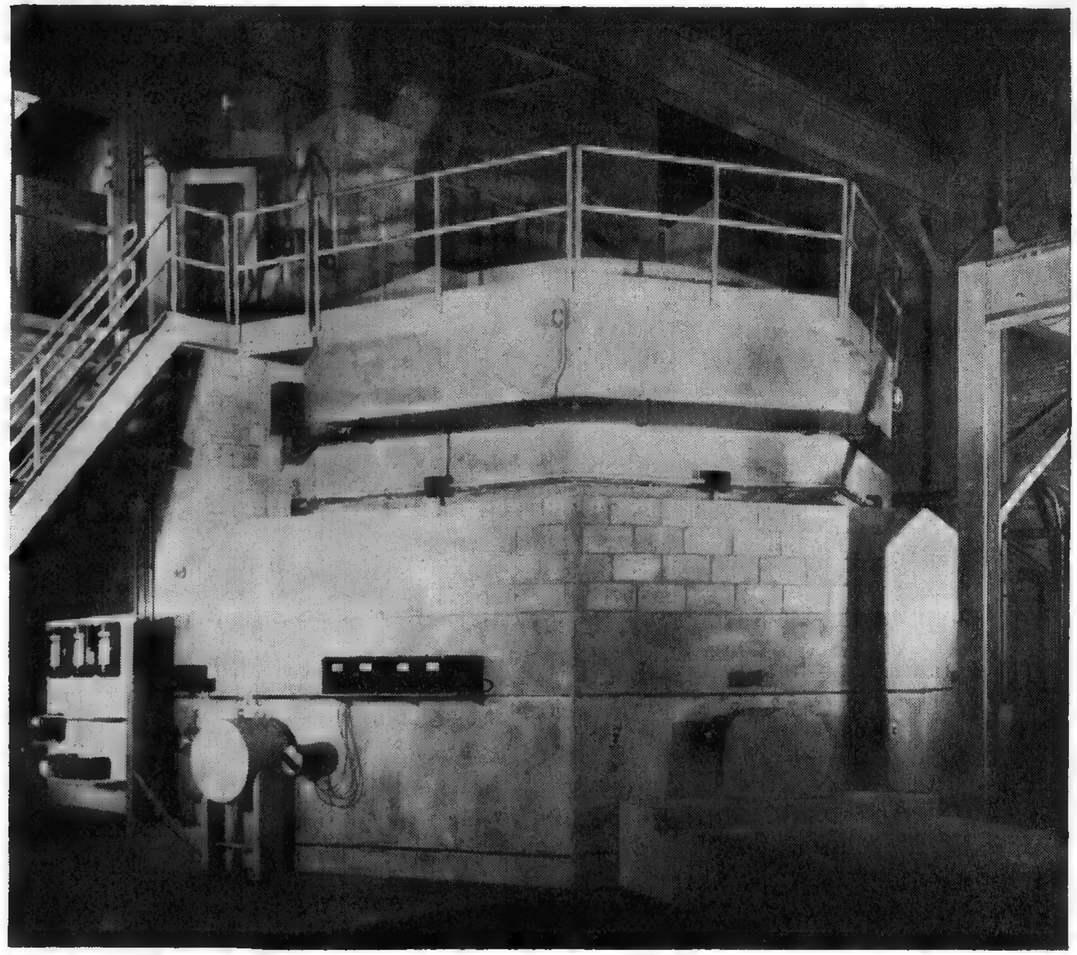


FIG. 13.28 General view of the Experimental Breeder Reactor at Arco, Idaho. (Argonne National Laboratory)

13.122. The coolant leaves the reactor at about 660°F and in the heat exchanger some of the heat is removed by a second loop of NaK; since this does not become radioactive, it does not require shielding. The heat exchanger in the second loop is the steam generator (boiler) in which superheated steam is produced at 550°F and 400 psig. The steam is used to drive a turbogenerator capable of producing about 200 kw of electricity. The first known generation of electricity from nuclear power was realized from the EBR in late December, 1951.

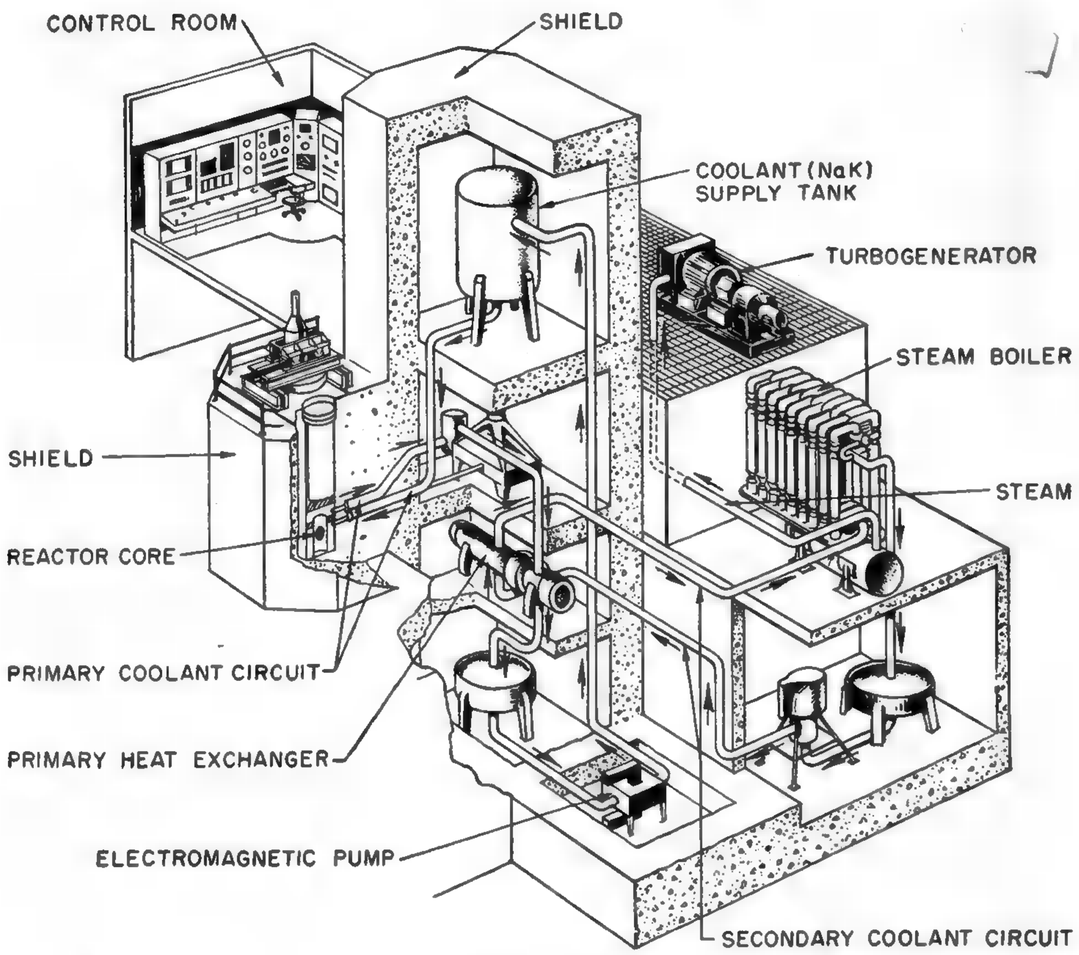


FIG. 13.29 Schematic representation of EBR and associated power plant. (Argonne National Laboratory)

13.123. In order to remove the heat released by fission products after shut-down, without the use of the regular heat-exchanger system, the primary NaK circulates by free convection through a finned-tube cooler at the base of a stack. As the air is drawn up the stack, it moves across the fins and thus cools the NaK. In this way, the core (after shut-down) can be cooled sufficiently without the use of an outside source of power.

13.124. In spite of its small size, the EBR has been able to operate at a power level of 1400 kw. The power density is at least 4 kw/in.^{3} or 250 kw/liter. It is believed that the power output was limited by auxiliary air cooling of some parts of the system. The average fast neutron flux is 2×10^{14} neutrons/(cm^2)(sec). However, since xenon poisoning is of no significance in a fast reactor, this flux presents no such problems as it might for a thermal reactor.

13.125. After the EBR had been operated for some time, analysis of samples of the core and the breeding blanket showed that the conversion ratio was at least unity, so that a power-breeder, operating with fast neutrons, is feasible. Based upon the success of this experiment, a new EBR-2 is being developed. This will ultimately use plutonium in the core, so that it will be a true breeder, although initially uranium-235 may be included for metallurgical reasons. The reactor will have a gross thermal power, in core and blanket, of 62,500 kw, and the associated turbogenerator will have a net electrical output of, at least, 15,000 kw. Liquid sodium, rather than NaK, will be the coolant. It is hoped that a specific power of 500 to 1500 kw/kg of fissionable material will be attained, since values of this order of magnitude are desirable if breeding is to occur at a reasonable rate (§ 1.157).

13.126. Pumps, heat exchangers, valves, and other hardware for the EBR-2 will be of a size suitable for large-scale application. The temperature of the sodium coolant delivered to the steam generator will be limited only by corrosion of the available materials. The steam temperatures and efficiencies attained should be comparable with those in modern central-station power plants.

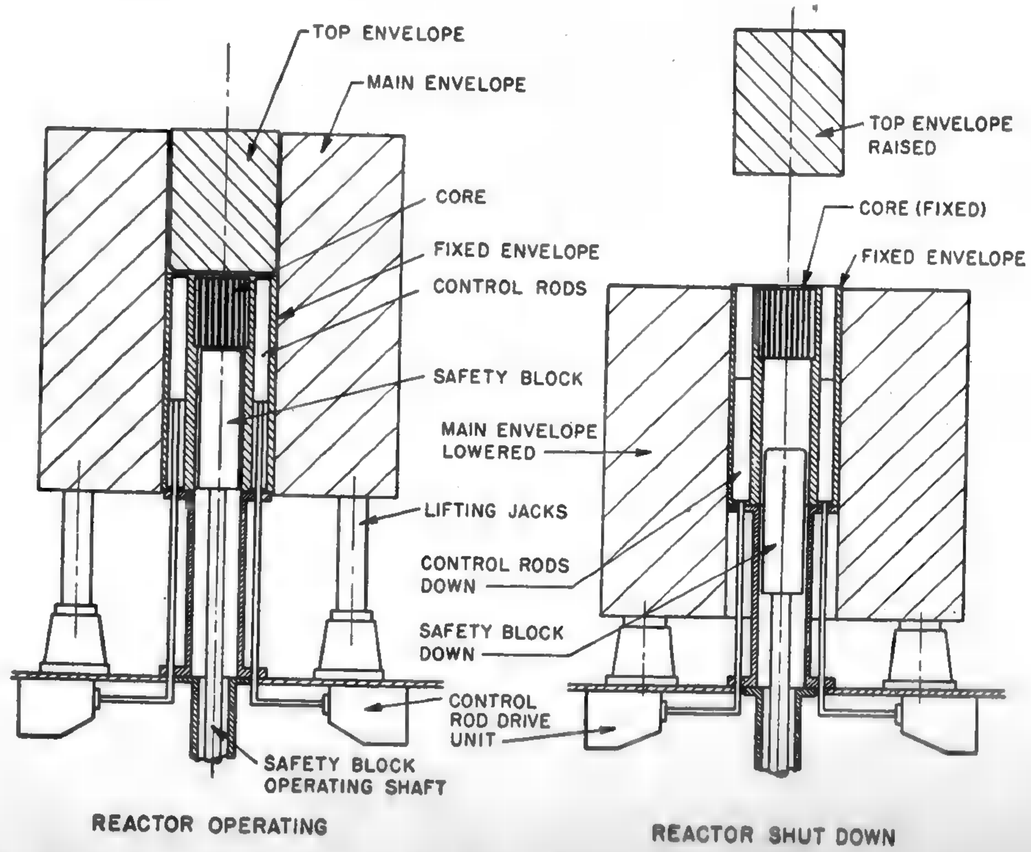


FIG. 13.30 Schematic representation of Zero Energy Fast Reactor at Harwell, England (Atomic Energy Research Establishment, Harwell, England)

*British Zero Energy Fast Reactor**

13.127. The Zero Energy Fast Reactor (ZEPHYR) at Harwell, England, is an experimental facility of low power for the study of the behavior of fast-neutron systems. It first became critical in February, 1954. There is no provision for cooling, so that the maximum operating level is usually 1 or 2 watts. The core is a cylinder, about 6 in. in diameter and 6 in. high, containing plutonium and natural uranium; the reflector blanket is also of natural uranium. Control is achieved, as it was in the Los Alamos Fast Reactor, by the movement of uranium rods in the reflector near the core (Fig. 13.30).

13.128. There is also a large movable block of uranium under the core; when this is pulled out the reactor is shut down. The lower end of this block is attached to a piston inside a cylinder; pressurized nitrogen is in the space above the piston, while the region below contains oil. A valve held in place by an electromagnet keeps the oil in the cylinder, but, if the magnet current is interrupted, the oil is released and the compressed nitrogen forces down the piston (and attached uranium block) at high speed. Return of the block to the operating position is achieved by closing the valve and applying pressure to the oil.

13.129. The ZEPHYR is located in a small room with concrete walls 2 ft thick and a removable concrete roof $1\frac{1}{2}$ ft thick, which serve as the shield. Provided the reactor is in a subcritical condition, the room can be entered and adjustments made to the reactor and apparatus. By means of a series of interlocks, start-up is prevented until the room is vacated and locked, and other necessary precautions are taken to avoid an accident. One of the purposes of the ZEPHYR is to obtain information for use in designing a fast-neutron breeder reactor for power production.

* L. R. Shepherd, *et al.*, *Chem. Eng. Prog. Symposium Series*, No. 13, **50**, 1 (1954); see also, *Nucleonics*, **12**, No. 3, 61 (1954).

APPENDIX

TABLE A.1. CONVERSION FACTORS

Multiply	*By*	*To Obtain*
Length		
centimeters	0.03281	feet
centimeters	0.3937	inches
feet	30.48	centimeters
inches	2.540	centimeters
Volume		
cubic centimeters	3.532×10^{-5}	cubic feet
cubic centimeters	0.06102	cubic inches
cubic feet	2.832×10^{4}	cubic centimeters
cubic feet	7.481	gallons
cubic feet	28.32	liters
cubic inches	16.39	cubic centimeters
gallons	0.1337	cubic feet
gallons	3.785	liters
liters	0.03532	cubic feet
liters	0.2642	gallons
Mass		
grams	2.205×10^{-3}	pounds
kilograms	2.205	pounds
pounds	453.5	grams
pounds	0.4535	kilograms
pounds	5×10^{-4}	tons (short)
tons (short)	2000	pounds
Heat and Energy		
British thermal units	251.8	calories
British thermal units	3.930×10^{-4}	horsepower hours
British thermal units	2.931×10^{-4}	kilowatt hours
British thermal units	0.2931	watt hours
calories	3.968×10^{-3}	British thermal units
electron volts	1.603×10^{-12}	ergs
electron volts	1.603×10^{-19}	watt seconds
horsepower hours	2544	British thermal units
kilowatt hours	3413	British thermal units
million electron volts	1.603×10^{-6}	ergs
million electron volts	1.603×10^{-13}	watt seconds
million electron volts	1.520×10^{-16}	British thermal units
watt hours	3.413	British thermal units

TABLE A.1. CONVERSION FACTORS (continued)

Multiply	*By*	*To Obtain*
Power		
Btu/hour	2.931×10^{-4}	kilowatts
Btu/hour	0.2931	watts
Btu/hour	3.930×10^{-4}	horsepower
horsepower	2544	Btu/hour
horsepower	0.7457	kilowatts
kilowatts	3413	Btu/hour
kilowatts	1.341	horsepower
watts	3.413	Btu/hour
Specific Heat		
Btu/(pound)(°F)	1.000	calories/(gram)(°C)
calories/(gram)(°C)	1.000	Btu/(pound)(°F)
Thermal Conductivity		
Btu/(lb)(ft²)(°F/ft)	4.134×10^{-3}	calories/(gram)(cm²)(°C/cm)
calories/(gram)(cm²)(°C/cm)	241.9	Btu/(lb)(ft²)(°F/ft)
Density		
cubic centimeters/gram	62.42	cubic feet/pound
cubic feet/pound	0.01601	cubic centimeters/gram
Viscosity		
centipoise	2.419	pounds/(hour)(foot)
poise	241.9	pounds/(hour)(foot)
pounds/(hour)(foot)	0.4134	centipoise
pounds/(hour)(foot)	4.134×10^{-3}	poise

TABLE A.2.* MICROSCOPIC THERMAL-NEUTRON CROSS SECTIONS OF WHOLE ELEMENTS

Element	At. No.	Chemical At. Wt.	Absorption (barns)	Scattering (barns)
Hydrogen	1	1.0080	0.33	20–80
Deuterium	1	2.015	0.00046	5.4
Helium	2	4.003	v. small	0.8
Lithium	3	6.94	70	1.4
Beryllium	4	9.01	0.009	7
Boron	5	10.82	750	4
Carbon	6	12.01	0.0045	4.8
Nitrogen	7	14.008	1.8	10
Oxygen	8	16.000	0.0002	4.2
Fluorine	9	19.00	0.009	4.1
Neon	10	20.18	2.8	2.4
Sodium	11	22.997	0.50	4.0
Magnesium	12	24.32	0.060	3.6
Aluminum	13	26.98	0.21	1.4
Silicon	14	28.09	0.13	1.7
Phosphorus	15	30.98	0.20	5
Sulfur	16	32.07	0.49	1.1
Chlorine	17	35.457	31.6	—
Argon	18	39.94	0.62	1.5
Potassium	19	39.100	2.0	1.5
Calcium	20	40.08	0.43	9
Scandium	21	45.10	23	—
Titanium	22	47.90	5.6	4
Vanadium	23	50.95	5.1	5
Chromium	24	52.01	2.9	3.0
Manganese	25	54.93	13	2.3
Iron	26	55.85	2.4	11
Cobalt	27	58.94	37	5
Nickel	28	58.69	4.5	17.5
Copper	29	63.54	3.6	7.2
Zinc	30	65.38	1.1	3.6
Gallium	31	69.72	2.7	4
Germanium	32	72.60	2.3	3
Arsenic	33	74.91	4.1	6
Selenium	34	78.96	12	13
Bromine	35	79.916	6.5	6
Krypton	36	83.8	28	7.2
Rubidium	37	85.48	0.70	12
Strontium	38	87.63	1.2	10
Yttrium	39	88.92	1.4	3
Zirconium	40	91.22	0.18	8
Niobium or Columbium	41	92.91	1.1	5
Molybdenum	42	95.95	2.4	7
Technetium	43	—	—	—
Ruthenium	44	101.7	2.5	6
Rhodium	45	102.9	150	6
Palladium	46	106.7	8	3.6
Silver	47	107.88	60	6

* Most of the data for this table were supplied by H. Pomerance and W. H. Sullivan.

TABLE A.2.* MICROSCOPIC THERMAL-NEUTRON CROSS SECTIONS OF WHOLE ELEMENTS (continued)

Element	At. No.	Chemical At. Wt.	Absorption (barns)	Scattering (barns)
Cadmium	48	112.41	2400	7
Indium	49	114.76	190	2.2
Tin	50	118.70	0.65	4
Antimony	51	121.76	5.5	4.3
Tellurium	52	127.61	4.5	5
Iodine	53	126.91	6.7	3.6
Xenon	54	131.3	35	4.3
Cesium	55	132.91	29	20
Barium	56	137.36	1.2	8
Lanthanum	57	138.92	8.9	18
Cerium	58	140.13	0.70	9
Praseodymium	59	140.92	11	—
Neodymium	60	144.27	44	25
Promethium	61	—	—	—
Samarium	62	150.43	6500	—
Europium	63	152.0	4500	—
Gadolinium	64	156.9	44,000	—
Terbium	65	159.2	44	—
Dysprosium	66	162.46	1100	—
Holmium	67	164.94	64	—
Erbium	68	167.2	166	—
Thulium	69	169.4	118	—
Ytterbium	70	173.0	36	12
Lutetium	71	174.99	108	—
Hafnium	72	178.6	115	—
Tantalum	73	108.88	21	5
Tungsten	74	183.92	19	5
Rhenium	75	186.31	84	14
Osmium	76	190.2	15	11
Indium	77	193.1	440	—
Platinum	78	195.23	8.1	10
Gold	79	197.2	98	9.3
Mercury	80	200.61	380	20
Thallium	81	204.39	3.3	14
Lead	82	207.21	0.17	11
Bismuth	83	209.00	0.032	9
Polonium	84	—	—	—
Astatine	85	—	—	—
Radon	86	—	—	—
Francium	87	—	—	—
Radium	88	—	—	—
Actinium	89	—	—	—
Thorium	90	232.12	7.0	13
Protactinium	91	—	—	—
Uranium	92	238.07	7.42	8.2

TABLE A.3.* PROPERTIES OF SOME SOLID MATERIALS USED IN REACTORS

Material	Density lb/ft³ ρ	Density °F	Specific Heat Btu/(lb)(°F) c_p	Specific Heat °F	Coefficient of Thermal Expansion per °F × 10⁶ α	Coefficient of Thermal Expansion °F	Thermal Conductivity Btu/(hr)(ft²)(°F/ft) k	Thermal Conductivity °F	Young's Modulus 10⁶ lb/in.² E	Young's Modulus °F	Poisson's Ratio ν	Poisson's Ratio °F	Tensile Strength lb/in.² Strength	Tensile Strength °F
Aluminum (2S)	169	68	0.222	32	13.0	68–212	128	77	10	75	0.33	75	13,000	75
			0.230	212	13.7	68–392			9.5	300			7500	300
			0.237	392	14.2	68–572			8.5	500			3500	500
			0.245	572					4.0	700			1500	700
Beryllium (Extruded flake)	115	77	0.473	77	8.3	200	80	200	39–43	~75	0.024	~75	62,000 (ult.)	212
	110	1832	0.506	261	9.4	400	73	400					53,000	400
			0.570	621	10.0	600	68	600					41,000	600
			0.635	981	10.5	800	61	800					27,000	800
			0.699	1341	11.1	1000								
Beryllium oxide (Varying densities)	137–175	75	0.219	32	3.1	77–212	29	400	39	77–1470	0.34	77–1470	15,000	77–750
			0.308	212	5.3	77–1112	22	600	38	1830	0.35	1830	13,500	1470
			0.420	752	6.0	77–1832	18	800	28	2010			10,500	1830
			0.492	1472					10	2190			8000	2010
Graphite (Mainly commercial blocks)	100	32	1.72	77	4.0	32	76	600	1.1	0			3100	0
	105	392	2.38	261	4.5	482	65	800	1.2	1000			3300	1000
	100	1112	3.36	621	4.9	932	59	1000	1.25	1500			3600	2000
	93	2192	4.28	1701	5.3	1382			1.3	2000			4900	4000
			4.72	2061									6300 (max.)	4700
Stainless steel (Type 347)	498	75	0.12	75	9.2	212	9.0	212	29	75	0.28	75	4000	5300
					10.0	1112	12.6	932	28	300	0.29	300	85,000	75
					11.1	1832			26	500	0.30	500	74,500	300
									24	800	0.31	800	69,000	500
									22	1100	0.32	1100	75,500	800
Thorium	730	75	0.0284	211	6.4	86–212	22	212	10	75	0.25	75	56,000	1100
			0.0285	390	6.6	86–932	26	1202	8.7	600			31,200 (ult.)	75
					6.9	86–1832			7.0	1000			16,500 (yield)	200
													12,000 "	400
Uranium (cast)	1155	75	0.0279	81	12	77–257	15	200					9500 "	600
			0.0320	441	15	77–617	17.5	400					3000 "	1000
			0.0376	801	20	77–1202	18	600						
			0.0467	1161										
Uranium (22 wt. %)–aluminum alloy	~200	75			11.1	68–212	97	400	11	75			29,000	75
					11.8	69–392	94	600						
					12.2	68–572	92	750						
					12.6	68–932								
Zirconium (Crystal bar)	406	75	0.0692	75	3.42 (C-axis)	77	11.8	212	14	75	0.32–0.35	75	18,600	75
	397	1585	0.0739	260	3.16 (A-axis)	77	11.4	392	10.5	600			19,000 (17% U)	75
			0.0815	800									14,000 "	300
			0.0862	1340									8100 "	570
													15,600	100
													31,000	200
													26,500	400
													21,000	600
													15,600	

* Most of the data for this table and the next were supplied by the Reactor Experimental Engineering Division, Oak Ridge National Laboratory.

TABLE A.4. PROPERTIES OF REACTOR COOLANTS

Material	Density lb/ft³ ρ	°F	Specific Heat Btu/(lb)(°F) c_p	°F	Viscosity lb/(hr)(ft) μ	°F	Thermal Conductivity Btu/(hr)(ft²)(°F/ft) k	°F
Helium (1 atm)	0.01191	0	1.240	all temps.	4.58×10^{-2}	45	8.30×10^{-2}	45
	0.0087	212			5.81	405	10.1	242
					7.63	765	11.7	441
					9.26	1125	12.5	548
					10.76	1485	18.2	800
							16.7	1000
Dry air (1 atm)	0.08624	0	0.240	45	4.23×10^{-2}	45	1.43×10^{-2}	45
	0.059	212	0.242	243	5.43	243	1.90	243
	0.038	570	0.246	441	6.46	441	2.34	441
			0.252	639	7.38	639	2.73	639
			0.270	1250	8.21	1250	3.09	1250
			0.287	2060	9.74	2060	3.77	2060
Carbon dioxide (1 atm)	0.093	212	0.22	212	3.8×10^{-2}	100	1.2×10^{-2}	212
	0.059	570	0.23	570			2.0	570
Water vapor (500 psia)	1.01	500	0.68	467	4.6×10^{-2}	500	2.7×10^{-2}	467
	0.863	600	0.41	600	5.0	600	2.9	600
	0.767	700	0.35	700	5.5	700	3.0	700
	0.694	800	0.34	800	6.1	800	3.2	800
	- 0.636	900			7.3	900	4.0	900
Water (saturated liquid)	61.8	100	0.997	100	1.66	100	0.363	100
	60.1	200	1.00	200	0.75	200	0.395	200
	53.6	400	1.06	400	0.35	400	0.372	400
	42.2	600	1.55	600	0.20	600	0.285	600
Sodium	57.9	212	0.331	212	1.66	219	49.7	212
	55.6	482	0.320	392	1.22	334	54.4	392
	53.3	752	0.306	752	0.922	482	43.8	572
	51.0	1022	0.300	1112	0.651	752	41.2	752
	48.7	1292	0.303	1472	0.440	1112	38.6	932
Sodium (22 wt %) –potassium alloy (NaK)	55.3	212	0.269	212	14.9	212	1.32	219
	53.1	482	0.255	572	15.3	392	0.997	334
	50.8	752	0.249	932	15.7	572	0.765	482
	48.6	1022	0.248	1112	16.0	752	0.557	752
	46.3	1292	0.253	1472	16.3	932	0.390	1292
Lithium	31.7	392	1.40	392	1.43	362	2.2	424–451
	30.6	752	1.23	572	1.39	382		
	29.6	1112	1.09	752	1.34	406		
	28.5	1472	1.00	932	1.19	484		
	27.5	1832			1.10	546		
Bismuth	625	600	0.0345	600	3.92	600	9.5	600
	608	1000	0.0369	1000	2.66	1000	9.0	1000
Lead (44.5 wt %) –bismuth eutectic	653	392	0.035	291–676	4.11	630	5.3	320
	636	752			3.34	842	5.6	392
	619	1112			3.12	932	5.8	464
	602	1472			2.98	1022	6.3	572
	584	1832			2.83	1112	6.5	608
Sodium hydroxide	110	700	0.540	700	7.9	700	1.2	700
	108	800	0.508	800	5.9	800	1.07	800
	106	900	0.476	900	4.7	900	0.88	900
	104	1000	0.443	1000	3.8	1000	0.71	1000
	103	1100	0.410	1100	3.1	1100	0.55	1100

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